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CO-DISPOSAL OF LOW-LEVEL RADIOACTIVE WASTE WITHIN SANITARY LANDFILLS

FINAL REPORT

October 1981

by

E. S. K. Chian, F. G. Pohland, B. Kahn,
K. C. Chang, L. Roland, W. H. Cross

Georgia Institute of Technology
Atlanta, Georgia 30332

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Two lab-scale landfill units receiving municipal refuse were constructed and spiked with low-level radioactive isotopes (i.e., Co-58, Sr-85, Cs-134 and H-3). They were operated under leachate recycle and non-recycle modes to assess the relative rates of stabilization of these landfill units, together with the fate of the radioactive isotopes within these systems. Weekly samples of leachate and gas were collected and analyzed for gross parameters (e.g., TOC, COD, BOD, etc.), specific organic compounds, metals, radioactive isotopes,

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20. Abstract (continued)

gas composition and gas production rates.

It was shown in this study that concentrations of Co-58 were reduced greatly in the leachate after six months of operation, whereas Sr-85 and Cs-134 were reduced to a some what lesser extent. The activity of H-3 was, however, detected in the gas generated from the landfill units. The mechanisms of removal of these radioactive isotopes were proposed and discussed in some detail. Results of this study also conform with reports that leachate recycle indeed accelerates the rate of landfill stabilization processes.

It is believed that information provided by this study will be useful in leading toward a rational design of pilot-scale landfill units to explore fully the potential of co-disposal of low-level radioactive wastes within the sanitary landfills.

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1.1 Problem Description

The disposal of low-level radioactive waste (LLRW) by shallow land burial methods has been used for more than three decades in the United States. According to the past experience, the future expansion of nuclear power plants will generate four significant problem areas.

1.1.1 Migration of Radioactive Waste

During the past, some of the burial sites for radioactive material have had leakage problems caused by the groundwater and rainfall entering the disposal site (5,20,32,41). Such water may slowly leach and carry radioactive materials through the water bearing strata or fissures. Therefore, some of the radioactive material may reappear in surface waters and underground aquifers.

1.1.2 Limited Land Burial Space

There are six commercially operated burial sites for radioactive materials in the United States. The sites comprise a licensed usable area of 358 acres. At an average volume of 7,700 cubic meter of packaged waste buried per acre of land, it is estimated that 2.9 million cubic meter of waste can be buried. Assuming a moderate nuclear industry growth and projected waste generation rates, it has been calculated that the entire licensed land burial capacity will be depleted by 1988 (1). Currently, in the eastern portion of the United States, one site is not operating (Sheffield, IL), one is nearly filled to licensed capacity (Barnwell, SC), and another two have experienced migration of radioactive materials (West

Valley, NY and Maxey Flats, KY). Therefore, the eastern U. S. burial sites could become filled or unavailable sooner than is projected.

1.1.3 Transportation Risk

The projected growth rate of nuclear power indicates that 90 percent of the total nuclear electrical generating capacity will be constructed in the eastern portion of the United States (1). Therefore, more LLRW will be generated near eastern burial sites, and a limited licensed capacity in this area will force many eastern utilites to ship their LLRW to more distant western sites thereby incurring higher transportation costs.

Besides transportation costs, there is also a high potential of incidental radioactive contamination risk caused by vehicles transporting the LLRW across the continent from the east to the west coast.

1.1.4 Energy Consideration

Transporting large amounts of LLRW to burial sites will require a considerable amount of energy. Energy can be consumed by the transport vehicles and the processes that are needed to fulfill the regulations issued by the Department of Transportation and by the Nuclear Regulatory Commission (61) for the safe transport of radioactive waste.

The co-disposal of LLRW with municipal solid wastes in sanitary landfills is considered an alternative solution to the above problems, since the infiltration by rainfall can be controlled, the leachate generated from the landfill can be collected for subsequent monitoring and/or treatment, and the landfill gases can be accumulated and used for fuels. This approach would greatly reduce the probability of leachate contamination of the surrounding environment over an extended and often indeterminant period of time.

1.2 Objectives of Research

The objectives of this study were to investigate containment of LLRW in sanitary landfills and associated attenuation mechanisms. Simulated landfill units were constructed for this study. Leachate and gas analyses were performed on these landfill units.

1.2.1 Simulated Landfill Units

Laboratory landfill units were constructed to investigate the containment of radionuclides in leachate under the influence of simulated rainfall events with and without leachate recycle. The simulated landfill units were filled with LLRW and municipal waste without leachate recirculation, or with LLRW and municipal waste with leachate recirculation (see Figures 7 and 8).

The proposed radioactive isotopes used in this study were Co-60, Cs-137, and Sr-90, because they are the common constituents presented in low-level radioactive waste (5, 65). Due to the findings by Pohland (48), when applying leachate recycle technique to a landfill, it will reach stabilization within months, hence short-life radioactive isotopes of Co-58, Cs-134, and Sr-85 were selected for Co-60, Cs-137, and Sr-90, respectively. Tritium was used as a tracer of water for the determination of water migration and its role in biological metabolic cycle.

1.2.2 Leachate Analysis

The gross parameters of leachate, such as 5-day biochemical oxygen demand (BOD_5), chemical oxygen demand (COD), total organic carbon (TOC), pH, conductivity, oxidation reduction potential (ORP), suspended solids, volatile solids, and alkalinity, etc., were analyzed according to procedures outlined in Standard Methods (57) and Compilation of Methodology Used for Measuring Pollution Parameters of Sanitary Landfill

Leachate (8). These were followed by extensive analysis of the organic matter and heavy metals in the leachate. The leachate was concentrated and fractionated by means of membrane fractionation, gel permeation and followed by functional group analyses (such as carboxylic, phenolic, hydroxide, carbohydrates and proteins). Direct injection of aqueous solution or acid extract of leachates were used with gas chromatographic analyses. Quality assurance and quality control procedures were followed in these analyses.

1.2.3 Gas Analysis

The gas generated from the lab-scale landfill units was collected and analyzed for constituents including the radionuclides that might appear in the gas, i.e., H_2^3O, and CH_4^3. Water vapor, potentially containing tritium, was extracted by condensing it in a cold trap, the remaining gaseous methane was combusted in an oxygen atmosphere and the resulting water was collected in a second cold trap (5). Other analyses of gas composition using a molecular sieve adsorption was also included in the investigations.

1.3 Literature Review

1.3.1 Low-Level Radioactive Wastes

1.3.1.1 Sources and Quantity

The activities that begin with uranium ore and end with the production of nuclear fuel and radioactive wastes involve the "nuclear fuel cycle" which is then the source of a large fraction of the radioactive wastes currently generated in the U. S. Other sources are medical, industrial research, and education applications of radionuclides (52, 65).

The fuel cycle consists of: (1) mining uranium ore; (2) milling the ore to produce uranium oxide; (3) enriching the uranium product; (4) fabricating the enriched uranium into fuel elements; (5) irradiating the fuel elements in a nuclear reactor; (6) processing the spend fuel to recover fissionable materials; (7) disposing of the reprocessed wastes or spent fuel. The nuclear fuel cycles with and without reprocessing of spent fuel, are shown schematically in Figures 1 and 2.

Radioactive wastes are classified according to the activity and nature of the contained radioactivity. Three different classifications exist: low level, high level, and transuranium contaminated wastes. The first two categories differ only ~~in~~ the degree of concentration of radioactivity. The third category was established because wastes contaminated with long-lived alpha emitters require special handling. Low-level wastes are generally defined as waste that contain less than 10 nCi (nano Curie) of long-lived α-radiation per gram and have γ-radiation low enough to require only minimal biological shielding and remote handling, or that average less than one curie of activity per cubic foot of material or less than 200 mrem/hr. The yearly total volume of commerical low-level wastes buried within the United States have increased rapidly since the early 1960s (see Figure 3). The U. S. EPA has estimated that by the year 2000, approximately 30 million cubic meter of such wastes will have been generated and will require proper disposal (18, 19, 25, 41).

The sources and characteristics of low-level wastes for each step of the nuclear fuel cycle are listed in Table 1 (5). Table 2 gives the quantities of radioactive wastes generated each year by one large light water reactor (32).

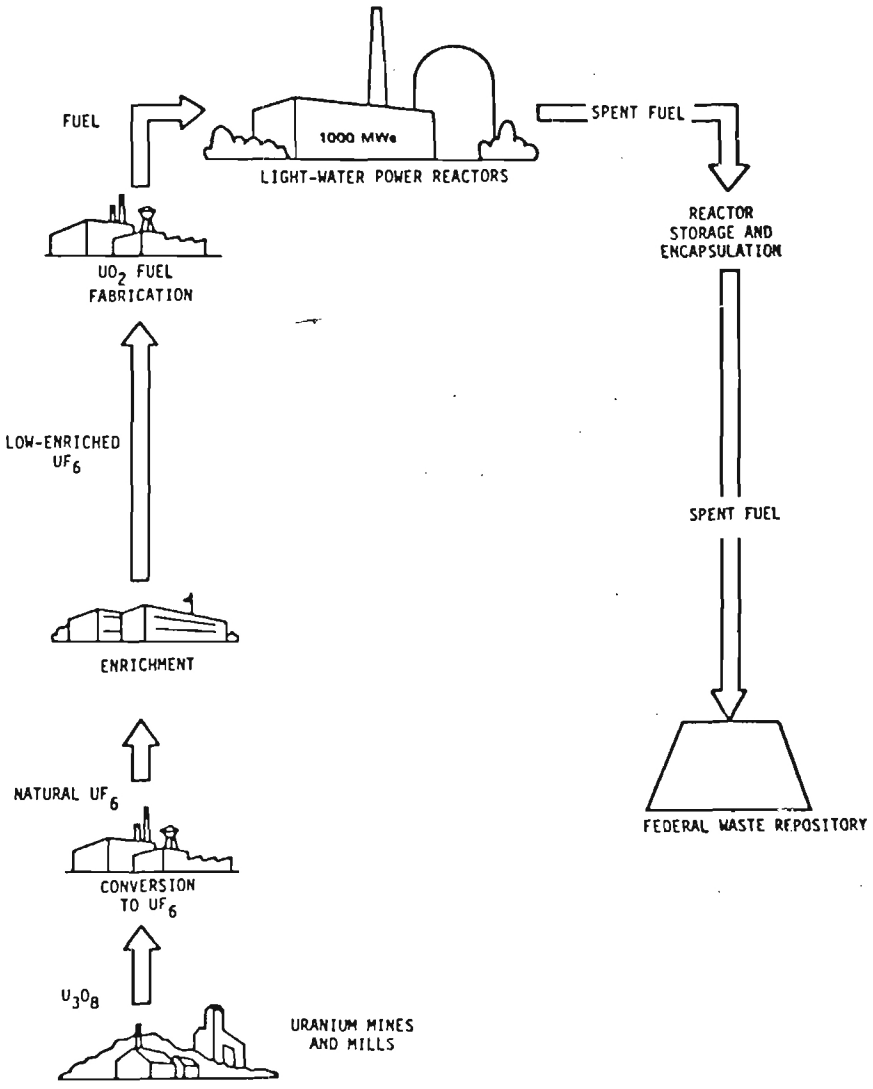


Figure 1. The Nuclear Fuel Cycle Without Reprocessing of Spent Fuel. (51)

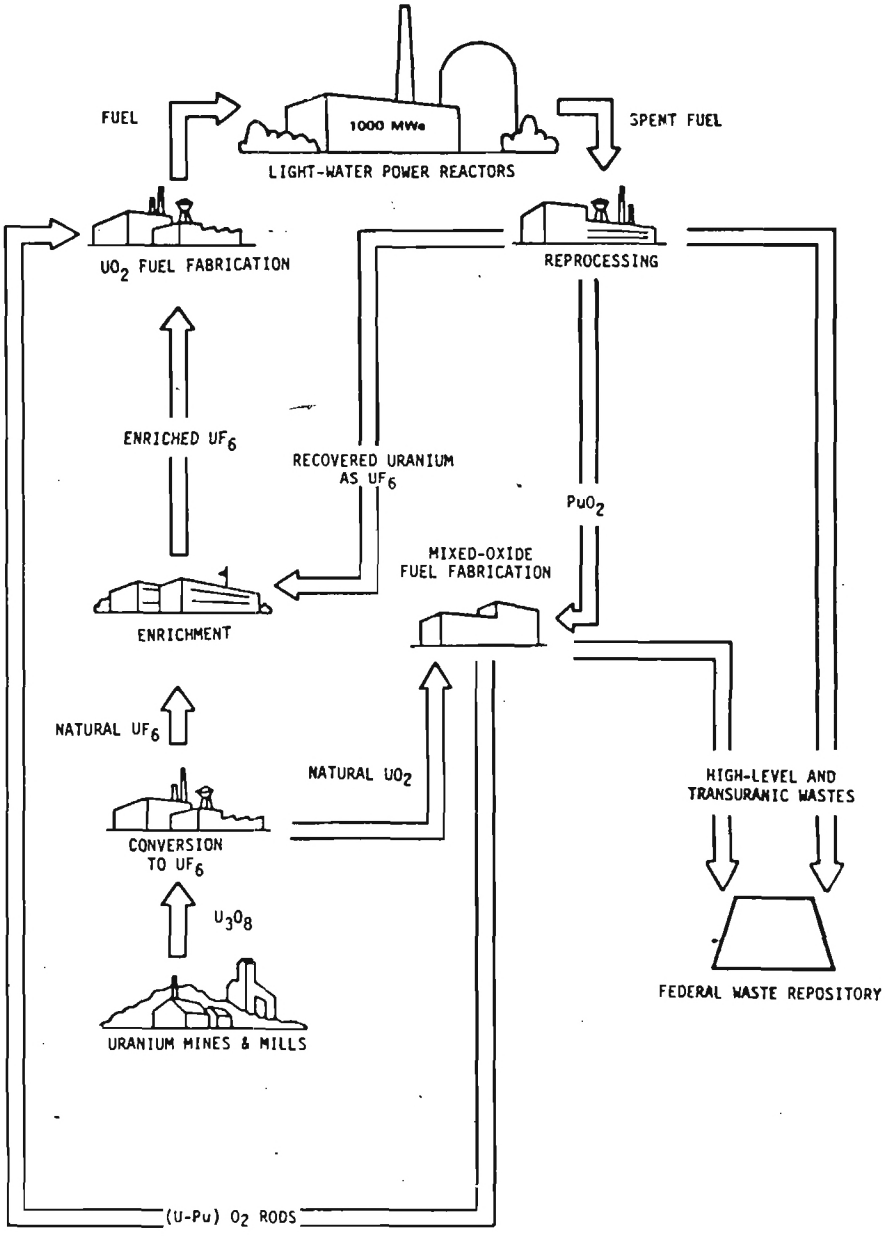


Figure 2. The Nuclear Fuel Cycle with Reprocessing. The Uranium and Plutonium Extracted from Spent Reactor Fuel are Recycled into New Fuel. (51)

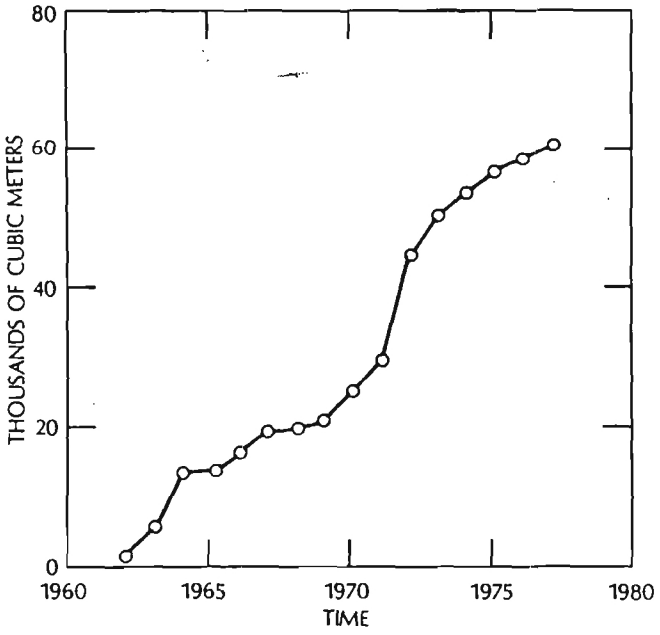


Figure 3. Yearly Total Volume of Commercial Low-Level Waste Buried Within the Conterminous United States. (41)

Table 1. Sources and Characteristics of Low-Level Non-Transuranic Wastes. (41)

Source	Original waste form	Major radioisotopes	Activity (Ci/MTHM[a])	Final waste form
1. Mining	Gaseous	^{222}Rn	0.46	Gaseous
2. Milling	Gaseous	^{222}Rn	1.97	Gaseous
	Airborne particulates	U, ^{226}Ra, ^{230}Th	1.31×10^{-4}	Airborne particulates
	Liquid	U, ^{226}Ra, ^{230}Th	1.30×10^{-2}	7.4×10^{5} liters of liquid waste per MTU[b]
	Solid	U, ^{226}Ra, ^{230}Th	4.15	317 m^3 of tailings (solid) per MTU
3. UF_6 conversion (fresh)	Airborne particulates	U	1.38×10^{-5}	Airborne particulates
	Liquid	U, ^{226}Ra, ^{230}Th	1.07×10^{-6}	1.3×10^{4} liters of liquid waste per MTU
	Solid	U, ^{226}Ra, ^{230}Th	9.40×10^{-3}	1.2 m^3 of solid waste (including CaF_2) per MTU
4. UF_6 conversion (recycle)	Airborne particulates	Fission products	8.73×10^{-6}	Airborne particulates
		^{238}Pu, ^{241}Pu	2.46×10^{-6}	
		U	3.32×10^{-5}	
	Solid	Fission products	0.57	~1.3 m^3 of solid waste (including CaF_2 and carbonate-leached ash) per MTU
		^{238}Pu, ^{241}Pu	4.12×10^{-5}	
		U	1.35×10^{-3}	
5. Enrichment (fresh)	Gaseous and airborne particulates	U	1.96×10^{-5}	Gaseous and airborne particulates
	Liquid	U	1.11×10^{-6}	675 liters of liquid waste per MTU

Table 1. (continued)

6.	Enrichment (fresh + 10% recycle)	Gaseous and airborne particulates	U	1.98×10^{-5}	Gases and airborne particulates
			^{237}Np	8.33×10^{-11}	
			^{239}Pu	1.67×10^{-13}	
			Fission products	2.29×10^{-4}	
		Liquid	U	1.16×10^{-6}	675 liters of liquid waste per MTU
			^{237}Np	1.67×10^{-9}	
			^{239}Pu	3.33×10^{-12}	
			Fission products	3.40×10^{-4}	
7.	Fuel fabrication (fresh + 10% recycle)	Airborne particulates	U, ^{234}Th, ^{234}Pa	5.60×10^{-6}	Airborne particulates
		Liquid	U, ^{234}Th, ^{234}Pa	1.20×10^{-3}	5.7×10^{5} liters of liquid waste per MTU
		Solid	U	7.34×10^{-3}	4.6 m^3 of solid waste (including CaF_2) per MTU
8.	Fuel fabrication (mixed oxide)	Airborne particulates	U	5.81×10^{-12}	Airborne particulates
			Pu, Am	2.08×10^{-7}	
		Solid	U, Pu, Am	3.47	~2265 (98.2) m^3 of solid waste per MT of plutonium (uranium) processed
9.	Reprocessing	Gaseous	^{3}H	436-526[c]	~0.008-0.01[c], ~0.01-0.013[d] m^3 of $Ca(OH)_2$ per MTHM
				544-665[d]	
		Gaseous	^{127}I, ^{129}I	0.655-0.936[c]	20 liters of liquid scrubber waste per MTHM; ~0.0011-0.0013[c], ~0.0016-0.0019[d] m^3 of concrete per MTHM
			^{131}I	0.686-0.992[d]	

Table 1. (continued)

9. (cont.)	Gaseous	^{85}Kr	7330-9100[c]	~0.05-0.06[c,d] cylinders (50 liters, 2200 psig) per MTHM
			5000-6020[d]	
	Gaseous	^{14}C	0.539-0.598[c]	$^{14}CO_2$
			0.293-0.314[d]	
	Solid	Pu, Am	0.53	~1586 (15.0) m^3 of solid waste per MT of plutonium (uranium) processed
		Fission products		

[a]Metric ton of heavy metal

[b]Metric ton of uranium

[c]First number is for BWR-U; second number is for PWR-U

[d]First number is for BWR-mixed oxide; second number is for PWR-mixed oxide

Table 2. Radioactive Wastes Generated Annually in the Fuel Cycle of a 1,000 Megawatt (Electric) Light Water Reactor with a 30 Metric Ton Fuel Off-Load.[a] (32)

Uranium mining and milling	96,000 metric tons of uranium mill tailings with 0.7 nanocuries of activity per gram 245,000 metric tons of tailings solutions Radon gas and daughters Low level radioactive dust and liquid effluents
Conversion to UF_6	1,200 cubic feet of low level solids, liquids, and sludge
Enrichment to 3-4 percent uranium-235	5,500 metric tons of enrichment tails containing 0.2 percent uranium-235 Low level solids: gaseous diffusion, 50 cubic feet; gaseous centrifuge, 2,900 cubic feet Low level liquids Airborne uranium (small quantities)
Fuel rod fabrication	750 cubic feet of low level solid wastes Process off-gases Low level liquids containing uranium, thorium, and protactinium
1,000 MWe Light Water Reactor	30 metric tons of spent fuel containing approximately 300 kilograms of U-235, 250 kilograms of plutonium isotopes, plus a variety of fission products; volume is 390 cubic feet Low level solids: boiling water reactor, 46,000 cubic feet with 38 millicuries of radioactivity per cubic foot; pressurized water reactor, 26,500 cubic feet with 43 millicuries of radioactivity per cubic foot Low level liquid effluents Reactor decontamination and decommissioning (only once during reactor lifetime): mothballing, 2,000 cubic feet; entombment, 80,000 cubic feet; dismantling and removal, 300,000-800,000 cubic feet
Reprocessing and mixed oxide fuel fabrication	Approximately 1.5 gallons of high level liquid waste per kilogram of spent fuel reprocessed, containing thousands of curies of radioactivity per gallon; volume is 240 cubic feet; 13.5 million curies 270 kilograms of plutonium; 3.8 million curies 530 cubic feet of spent fuel cladding hulls; 870,000 curies 200 cubic feet of low level wastes 1,060 cubic feet of transuranium-contaminated waste; 1.7 million curies
Spent fuel storage	6 cubic feet of low level solids and liquids Fission product gases
High level waste repository	Low level solids and liquids Transuranium-contaminated waste 0.6 to 1 acre required for disposal of 30 metric tons of spent fuel or waste equivalent

[a] 1,250 MWe reactor with 80 percent capacity factor; 1 metric ton = 1,000 kilograms = 2,200 pounds.

1.3.1.2 Nature and Hazard of Radioactivity

The spontaneous decay or disintegration of an unstable atomic nucleus, usually accompanied by the emission of ionizing radiation, is called "radioactivity." During radioactive decay, alpha particles (equal to a helium nucleus) and beta particles (equal to an electron) are emitted. Besides these, gamma rays and x-rays may also be produced; gamma-and x-rays are similar to light but of much greater energy.

Radioactive materials or radioactive nuclides impose a special hazard. Radioactive emissions are invisible, odorless, tasteless, and are able to penetrate matter; it is this property that makes them biologically unsafe. Unlike many chemical toxins that can be neutralized, radioactivity only disappears through natural decay which may take hundreds, thousands or even millions of years.

Radiation effects to living things can be short-term or long-term. Short-term effects, which can result from exposure to sufficiently intense radiation over a brief period of time, are characterized as "radiation sickness." Long-term effects may take years or decades to become apparent and can result from abrupt, large radiation exposure or extended exposure to low radiation levels. Table 3 lists the health effects of exposure to various levels of radiation (21, 32, 40).

1.3.1.3 Disposal Techniques

The safe disposal of radioactive wastes from the operation of nuclear power plants is considered one of the more urgent aspects of the nuclear fuel cycle that requires extensive evaluation because of the very long

Table 3. Biological Effects of Ionizing Radiation. (32)

Exposure Range	*Chronic Exposure*[a]	*Acute Exposure*[b]
Less than 1 rem	No observable effects; equivalent to exposure from background radiation for 5–10 years. Cancer risk: $1-2 \times 10^{-4}$ per rem for adult; greater than 4×10^{-4} per rem for fetus (may be as high as 6×10^{-4} to 6×10^{-3} per rem for children; 7×10^{-3} for adults).	No observable short-term effects.
1–50 rems	Chromosomal aberrations in blood. 0.3–30 leukemia cases per 10,000 person-rem observed in this exposure range; 0.5–1.2 thyroid cancers per 10,000 person-rem observed. Occupational exposure range.	Slight blood changes; decreased head circumference and increased leukemia risk in fetus.
50–100 rems	Approximate doubling dose for spontaneous mutations.	Mild symptoms of radiation sickness possible. Vomiting in 5 percent of those exposed to 100 rems within three hours.
100–200 rems	Approximate doubling dose for cancer.	Vomiting in 5 percent of those exposed to 100 rems to 50 percent of those exposed to 200 rems within three hours. Also, fatigue, loss of appetite, moderate blood changes that persist. Recovery within several weeks. Increased cancer risk; cataracts possible.
200–600 rems	Limited experience with regard to chronic exposure over 200 rems. Large increase in incidence of leukemia and other cancers. Uranium miners exposed to 700–1,000 rads, with maximum exposures estimated to be as great as 10,000 rads. Excess of lung cancer deaths may reach 600–1,100 in a population of 6,000 miners.	Vomiting: 50 percent at 200 rems within three hours; 100 percent above 300 rems within two hours. Also, loss of hair, other symptoms of radiation sickness. Death in 0–80 percent of those exposed within two months from hemorrhage and infection; recovery for survivors in one to twelve months.
600–1,000 rems		Vomiting within one hour, severe blood changes, hemorrhage, infection, loss of hair, damage to bone marrow. Death in 80–100 percent of those exposed within two months from hemorrhage and infection. Long convalescence for survivors.
1,000–3,000 rems		Vomiting within thirty minutes; radiation sickness. Gastrointestinal syndrome within five to fourteen days, including diarrhea, fever, severe blood changes, damage to bone marrow. Death in 90–100 percent of those exposed within two weeks due to circulatory collapse.
More than 3,000 rems		Vomiting within thirty minutes. Central nervous system syndrome within two days, including convulsions, tremor, loss of muscular control. Death in 100 percent of those exposed within one to forty-eight hours due to respiratory failure and brain edema.

[a] Exposure over extended time period.

[b] Exposure over period of twenty-four hours or less.

time scales involved. Disposal methods include: geological (17, 28, 51, 65), ocean (15,17, 22), ice (17, 33, 70) and space (17, 31, 42). Geological disposal (isolation) appears the most promising for all kinds of radioactive wastes; for low-level and/or transuranium wastes, shallow land burial techniques are used.

Although the general technical feasibility of several disposal concepts have been demonstrated, there is still a need for quantitative information on geological, geochemical and hydrological factors influencing the long-term stability of solidified wastes, and on the form and rate of migration of any dissolved or remobilized fractions of the radioactive material through subsurface aquifers.

It is highly probable that ground water and rainfall will reach many land disposal sites through seepage or direct discharge via thermal or mechanical fractures in the surrounding rock. Under these circumstances, waste materials will be leached and transported horizontally or vertically through water-bearing strata. Depending on flow rates and on intermediary sorption processes, some of the waste products may conceivably reappear in surface waters.

In considering migration of waste constituents from disposal sites, two types of transport can be proposed. One involves the movement of dissolved radioactive ions remaining in solution but subject to adsorption/desorption effects on exposed geologic surfaces. Such a transport process may lead to modified migration rates and may selectively retain those ions and complexes that are adsorbed most strongly. The other assumes that a finite fraction of the dissolved activity may adhere to very fine particulates and be carried along, subject primarily to surface forces between moving and stationary mineral surfaces and to competitive sorption effects between the water-particle and water-soil surface interfaces.

Besides the migration of radioactive wastes caused by aqueous leaching, other considerations including soil erosion in arid areas and earthquake should also be carefully examined before implementing land burial as a disposal technique. In general, desirable features of land burial sites for low-level radioactive wastes include (41):

1. a desert climate;
2. a deep groundwater table;
3. isolation from population centers
4. minimum surface erosion;
5. waste land not suitable for agriculture or recovery of potentially valuable mineral deposits;
6. good transportation access
7. availability of inexpensive and abundant construction materials
8. topography suitable for easy movement of heavy equipment, and
9. absence of any special environmental features such as scenic areas, unique flora or fauna, or high recreational potential.

A number of studies on the general hydrogeologic aspects of radioactive waste disposal have been reported during the last few years (6, 43, 69). In general, low level radioactive wastes have been produced from a variety of sources and have been disposed of at government-operated or commercially-owned sites. The 17 government-operated sites are those managed by the U. S. Department of Energy (DOE). Waste handling, storage, and burial operation are conducted within guidelines established earlier by the U. S. Atomic Energy Commission (59) and various locally derived guidelines. Materials that have been buried near the surface were classified as "solid radioactive waste other than solidified high level waste." They include paper, rags, rubber, synthetic-rubber-like materials, wood, glassware, carcasses, and excreta of experimental animals, protective

clothing, small pieces of contaminated equipment, and other contaminated matter. The locations, treatment amounts, and accumulations at each DOE sites are included on Figure 4 (26, 41) and in Tables 4-6 (41).

Land burial at commercial sites (or privately owned sites) is under the jurisdiction of the U. S. Nuclear Regulatory Commission (NRC) or an "Agreement State" (referring to an agreement between a qualified state and the U. S. AEC under Federal Statute 42 U.S.C.2021). The general guidelines for the application for a burial license and the necessary environmental statement are given in NRC Regulations 10 CFR 20, 30, 40, 51, and 70.

Locations of commercial burial sites, shown in Figure 4, are at West Valley, NY; Barnwell, SC; Morehead (or Maxey Flats) KY; Sheffield, IL; Beatty, NV; and Richland, WA. The commercial site at West Valley is operated by Chem-Nuclear Services, Inc. The remaining sites are operated by the Nuclear Engineering Company. The West Valley, NY and Maxey Flats, KY sites were closed because of off-site radioactive contamination and poor operating practices. The Sheffield, IL was closed because of an inability to acquire a new license that would allow site expansion. The remaining sites are still in operation. The total volume of solid radioactive wastes generated annually at commercial sites through 1975 is listed in Table 7 (62); estimated amounts of transuranium-contaminated wastes and amounts of plutonium for disposals by the year 2000 are listed in Table 8 (60).

During the past, disposal of low-level radioactive wastes was primarily by shallow land burial. Unfortunately, some burial sites have radioactive material leakage problems (5, 20, 66) which were caused by either surface flooding due to rainfall, or subsurface migration due to groundwater.

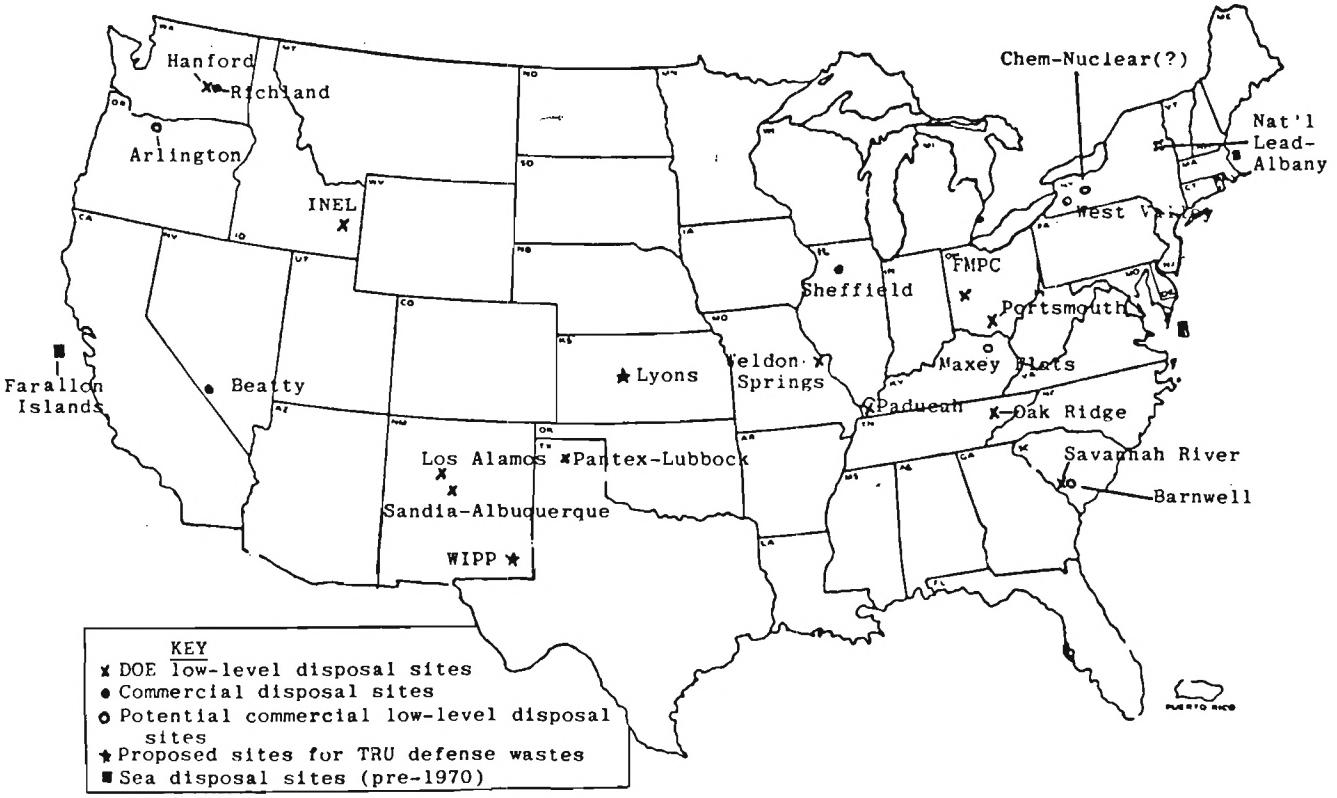


Figure 4. Low Level Waste Burial Sites in the United States. (32)

Table 4. Solid Radioactive Waste Stored and Buried at DOE (Previously ERDA) Sites Cumulative Through June 1974. (41)

Site	Hectares	(Acres)	Total Volume (cubic meters)	Kilocuries Buried	Kilocuries Remaining	Total TRU[a] (kilograms)	Total Uranium (kilograms)	Retrievable Storage	
								Cubic Meters	TRU[a] (kilograms)
Feed Materials Production Center (Fernald, Ohio)	2.9	(7.3)	73,000	1	1	–	2,483,000	–	–
Hanford Site (Richland, Washington)	98.4	(243.1)	200,000	1,870	900	365	592,100	4,300	40
Idaho National Engineering Laboratory (Idaho Falls, Idaho)	21.0	(52)	159,000	5,970	3,600	492	276,700	22,000	119
Lawrence Livermore Laboratory (Livermore, California)	2.3	(5.7)	700	[b]	[b]	–	32,900	–	–
Los Alamos Scientific Laboratory (Los Alamos, New Mexico)	23.1	(57.2)	220,000	210	160	15	245,100	700	8
National Lead Co. of Ohio (Niagara Falls, New York)	0.4	(1)	7,000	[c]	[c]	–	9,000	–	–
Nevada Test Site (Las Vegas, Nevada)	64.8	(160)	7,400	5	[b]	[c]	–	–	[b]
Oak Ridge Gaseous Diffusion Plant (Oak Ridge, Tennessee)	<0.4	(<1)	1,100	[c]	[b]	–	45,400	–	–
Oak Ridge National Laboratory (Oak Ridge, Tennessee)	23.8	(58.8)	176,000	<60	[b]	13	100	800	3
Oak Ridge Y-12 Plant (Oak Ridge, Tennessee)	2.1	(6.2)	26,000	[b]	[b]	–	NA	–	–
Paducah Gaseous Diffusion Plant (Paducah, Kentucky)	0.7	(1.7)	6,000	1	1	–	2,133,500	–	–
Pantex Plant (Amarillo, Texas)	<0.4	(<1)	100	[c]	[c]	2	20,200	[c]	2
Portsmouth Gaseous Diffusion Plant (Portsmouth, Ohio)	<0.4	(<1)	300	[c]	[b]	–	2,300	–	–
Sandia Laboratory (Albuquerque, New Mexico)	0.57	(1.4)	1,200	6	3	[c]	17,600	[c]	[b]
Savannah River Plant (Aiken, South Carolina)	37.6	(93)	273,000	8,440	4,280	50	72,000	2,000	44
Weldon Springs (St. Charles County, Missouri)	3.0	(2)	43,000	[c]	[c]	–	[b]	–	–
TOTAL	279	(690)	1,193,800	–	–	937	–	30,500	–

[a]TRU: Transuranium nuclides.
[b]Data not available.
[c]Negligible.

Table 5. Solid Radioactive Waste Stored and Buried at DOE (Previously ERDA) Sites During Fiscal Year 1974 (July 1, 1973, through June 30, 1974) (41)

Site	Total Quantities		Retrievable Storage	
	Cubic Meters	Kilocuries	Cubic Meters	TRU[a] (kilograms)
Feed Materials Production Center (Fernald, Ohio)	400	[c]	–	–
Hanford Site (Richland, Washington)	6,500	375	700	10
Idaho National Engineering Laboratory (Idaho Falls, Idaho)	7,100	57	3,800	36
Lawrence Livermore Laboratory (Livermore, California)	–	–	–	–
Los Alamos Scientific Laboratory (Los Alamos, New Mexico)	3,900	35	370	7
National Lead Co. of Ohio (Niagara Falls, New York)	–	–	–	–
Nevada Test Site (Las Vegas, Nevada)	580	[c]	[c]	[c]
Oak Ridge Gaseous Diffusion Plant (Oak Ridge, Tennessee)	10	[c]	–	–
Oak Ridge National Laboratory (Oak Ridge, Tennessee)	3,600	1	200	2
Oak Ridge Y-12 Plant (Oak Ridge, Tennessee)	90	[b]	–	–
Paducah Gaseous Diffusion Plant (Paducah, Kentucky)	90	[c]	–	–
Pantex Plant (Amarillo, Texas)	2	[c]	–	–
Portsmouth Gaseous Diffusion Plant (Portsmouth, Ohio)	7	[c]	–	–
Sandia Laboratory (Albuquerque, New Mexico)	80	<1	–	–
Savannah River Plant (Aiken, South Carolina)	14,100	175	80	2
Weldon Springs (St. Charles County, Missouri)	–	–	–	–
TOTAL	36,459	–	5,150	57

[a]TRU: Transuranium nuclides.
[b]Data not available.
[c]Negligible.

Table 6. Solid Radioactive Waste Generated at DOE (Previously ERDA) Facilities without Burial Grounds. (41)

Site	Cumulative Quantities through 1974				FY 1974[d] Quantities			
	Total (cubic meters)	Total (kilocuries)	TRU[a] Waste (cubic meters)	TRU[a] (kilograms)	Total (cubic meters)	Total (kilocuries)	TRU[a] Waste (cubic meters)	TRU[a] (kilograms)
Ames Laboratory (Ames, Iowa)	100	<1	–	–	10	[c]	–	1
Argonne/West (Idaho Falls, Idaho)	23,000	7	70	5	900	1	[c]	[c]
Atomics International (Santa Susana, California)	900	1	5	[c]	300	[c]	[c]	–
Bendix Plant (Kansas City, Missouri)	800	[c]	–	–	5	[c]	–	–
Bettis Atomic Power Laboratory (West Mifflin, Pennsylvania)	7,500	2	[c]	[c]	1,600	1	[c]	[c]
Brookhaven National Laboratory (Upton, L.I., New York)	300	45	–	–	200	45	–	–
Burlington ERDA Plant (Burlington, Iowa)	5	[c]	–	–	[c]	[c]	–	–
Fermi National Accelerator Laboratory (Weston, Illinois)	100	[c]	–	–	50	[c]	–	–
Lawrence Berkeley Laboratory (Berkeley, California)	200	1	40	[c]	60	1	[c]	–
Lovelace Foundation Laboratory (Albuquerque, New Mexico)	400	<1	1	[c]	110	[c]	–	[c]
Mound Laboratory (Miamisburg, Ohio)	24,000	85	17,000	5	1,500	12	1,000	<1
Naval Reactors Facility (Idaho Falls, Idaho)	17,000	3,200	1	–	900	10	–	–
Pinellas Plant (Clearwater, Florida)	700	110	–	–	60	12	–	–
Rocky Flats Plant (Golden, Colorado)	88,000	310	70,000	440	7,000	16	6,000	22
Shippingsport Atomic Power Station (Shippingsport, Pennsylvania)	700	[c]	–	–	300	[c]	–	–
TOTAL	163,705	3,760	–	450	12,995	–	7,000	–

[a]TRU: Transuranium nuclides.
[b]Data not available.
[c]Negligible.
[d]FY 1974: Fiscal Year 1974 (July 1, 1973, through June 30, 1974).

Table 7. Commercial Solid Radioactive Waste Generate Annually in the United States. (41)

Calendar Year	Volume (X 1,000 cubic meters)
1962	1.9
1963	6.3
1964	13.1
1965	13.1
1966	16.2
1967	19.4
1968	19.7
1969	21.4
1970	25.1
1971	29.6
1972	44.1
1973	50[a]
1974	54[a]
1975	57[a]

[a]Estimated by DOE (previously ERDA)

Table 8. Projected Annual Total Volume of Transuranium-Contaminated Solid Wastes and Amount of Plutonium for Disposal. (41)

	Volume (X 1,000 cubic meters)		Plutonium (kilograms)	
Calendar Year	ERDA[a]	Commercial[b]	ERDA[a]	Commercial[b]
1980	2	5	25	90
1990	1	10	25	600
2000	1	40	25	2,700

[a]Assuming a constant rate of generation of waste, and with a reduction in volume of up to 10:1 by suitable methods (incineration, compaction, etc.).

1.3.2 Sanitary Landfill

Based on past experience, land disposal in the form of a sanitary landfill has proven to be one of the most environmentally acceptable methods for the disposal of solid wastes. The term "sanitary landfill" has been defined in the ASCE Sanitary Landfill Manual (42) as "an engineered method of disposing of solid wastes on land in a manner that protects the environment, by spreading the waste in thin layers, compacting it to the smallest practical volume, and covering it with compacted soil by the end of each working day or, if necessary, more frequently."

After solid waste is placed in a sanitary landfill, it undergoes a number of simultaneous biological, physical, and chemical reactions. Among these reaction, the most important are: (1) biological decay of organic materials, either aerobically or anaerobically, with the releasing of gases and liquid; (2) chemical oxidation of materials; (3) movement of gases through the fill; (4) movement of liquids; (5) dissolving and leaching of organic and inorganic materials by water and leachate moving through the fill; (6) movement of dissolved materials by concentration gradient and osmosis; and, (7) uneven settlement caused by consolidation of material into voids (54).

Decomposition and stabilization in a landfill depend on many factors, such as the refuse quantity and composition, refuse placement characteristics, landfill depth, refuse moisture content, amount of oxygen content, the presence of inhibiting materials, the rate of water movement, and temperature. According to the difference between refuse types, and other interrelated influences, it is difficult to predict conditions that might exist in any landfill. In general, the rates of chemical and biological reactions in a sanitary landfill increase with the temperature and the moisture content in the fill (24, 58).

For a completed sanitary landfill, the stages of decomposition and stabilization of the refuse can be determined by examining the properties of leachate and gases from the landfill.

1.3.2.1 Landfill Leachate

Leachate can be attributed to many factors, including the water generated as a product of refuse decomposition. However, the main factor contributing to leachate quantity is inflow water from rainfall. After the landfill reaches field capacity, the leachate generation will continue on a cyclic pattern which will depend on the local rainfall, runoff, and evaporation patterns. Leachate quantities will correspond directly to the net infiltration rates.

Leachate carries many dissolved and suspended materials obtained during its travel through the refuse. The specific nature and concentration of the substances depends on the landfill refuse composition as well as its degradation stage. Typical concentration ranges are presented in Table 9 (2, 14, 58).

1.3.2.2 Gases in Landfill

Several gases are generated within landfills as a result of decomposition processes. The principal gases found in landfills include carbon dioxide, carbon monoxide, hydrogen, ammonia, hydrogen sulfide, methane, nitrogen, oxygen, and trace mercaptans. Data on the molecular weight and density of these gases are presented in Table 10.

During the earlier stages, refuse undergoes aerobic decomposition, and the main gas generated is carbon dioxide. Once all free oxygen is depleted, refuse decomposition becomes anaerobic, and the main gases become methane and carbon dioxide. Hydrogen sulfide is also formed occasionally in this stage. Typical data on the percentage distribution of gases found in a landfill are reported in Table 11.

Table 9. Data on the Composition of Leachate from Landfills. (58)

Constituent	Range mg/ℓ
BOD_5 (5-day biochemical oxygen demand)	2,000–30,000
TOC (total organic carbon)	1,500–20,000
COD (chemical oxygen demand)	3,000–45,000
Total suspended solids	200– 1,000
Organic nitrogen	10– 600
Ammonia nitrogen	10– 800
Nitrate	5– 40
Total phosphorus	1– 70
Ortho phosphorus	1– 50
Alkalinity as $CaCO_3$	1,000–10,000
pH	5.3– 8.5
Total hardness as $CaCO_3$	300–10,000
Calcium	200– 3,000
Magnesium	50– 1,500
Potassium	200– 2,000
Sodium	200– 2,000
Chloride	100– 3,000
Sulfate	100– 1,500
Total iron	50– 600

Table 10. Molecular Weight and Density of Gases Found in Sanitary Landfills at Standard Conditions (0°C, 1 atm). (58)

Gas	Formula	Molecular weight	Density	
			g/l	lb/ft^3
Air			1.2928	0.0808
Ammonia	NH_3	17.03	0.7708	0.0482
Carbon dioxide	CO_2	44.00	1.9768	0.1235
Carbon monoxide	CO	28.00	1.2501	0.0781
Hydrogen	H_2	2.016	0.0898	0.0056
Hydrogen sulfide	H_2S	34.08	1.5392	0.0961
Methane	CH_4	16.03	0.7167	0.0448
Nitrogen	N_2	28.02	1.2507	0.0782
Oxygen	O_2	32.00	1.4289	0.0892

Table 11. Typical Percentage Distribution of Landfill Gases During First 48 Months (58)

Time interval since start of cell completion, months	Average percent by volume		
	Nitrogen, N_2	Carbon dioxide, CO_2	Methane, CH_4
0–3	5.2	88	5
3–6	3.8	76	21
6–12	0.4	65	29
12–18	1.1	52	40
18–24	0.4	53	47
24–30	0.2	52	48
30–36	1.3	46	51
36–42	0.9	50	47
42–48	0.4	51	48

Carbon dioxide is of concern because it is water soluble, moreover, its density is higher than air so it tends to move downward in the form of liquid and gas phases. Once it reaches the ground water aquifer, it lowers the pH and increased the corrosiveness of ground water, which in turn increase the hardness and mineral content of ground water through solubilization (30).

Methane is virtually insoluble in water, but will move by diffusion and convention up through the refuse into the atmosphere or surrounding areas. When methane is present in air in concentrations between 5 and 15 percent, it is explosive. However, there is no oxygen in a landfill when methane concentrations in it reach this critical level so there is no danger that the fill will explode (2).

1.3.2.3 Leachate Recirculation

The concept of leachate recirculation to enhance the stabilization of a sanitary landfill has been developed and studied by Pohland (34, 47-50). It was found by Pohland (48) that by containing and recirculating leachate through a landfill, more rapid development of an active anaerobic bacterial population of methane formers, increasing rates and predictability of stabilization of readily available organic pollutants, dramatically decreasing times required for stabilization, and reduced potential for environmental impairment can be achieved. Moreover, leachate recirculation with pH control and sludge seeding further enhanced efficiency so that the time required for biological stabilization of the majority of organic pollutants in the leachate was reduced to a matter of months rather than years (48). Advantages also include a better operational control, energy recovery, rapid realization of potentials for land reclamation of ultimate use, and physical control over leachate residuals after such stabilization. Futhermore, it permits better consideration of ultimate disposal requirements,

i.e., direct discharge versus land disposal with or without further treatment (49). The removal of heavy metals in leachate was also enhanced by the increased stabilization rates, reducing conditions conducive to precipitate formation, and filtering action promoted by the recirculation of leachate through the landfill mass (50).

Based on the findings by Pohland et al. (48), leachate recycle should be used as an option for better design, operation, and control of landfill disposal sites.

II. MATERIALS AND METHODS

2.1 Materials

2.1.1 Solid Wastes

The solid waste was obtained from the Buford Highway Shredding Facility in DeKalb County, Georgia. The solid waste was mainly municipal type refuse. Bulky materials were removed prior to the shredding process. Each of two 208-liter (55-gal) polyethylene containers were filled with 55 kg (wet weight) of shredded municipal solid waste and the final volume of the solid waste after compaction was about 170 liters. The density of solid waste was 305 Kg/m^3 (wet weight) before adding water to attain field capacity.

Characterization of the solid waste included determination of moisture content, volatile solids content, calorific value, elemental composition (i.e., C.H.N. content), and nutrient requirements (e.g., NH_3, TKN, orthophosphate and total phosphate).

2.1.2 Radionuclides

One millicurie (mCi) each of Co-58, Cs-134, and Sr-85, and 0.25 mCi of H-3 were purchased from the New England Nuclear Company (Cambridge, MA). The procedures for the preparation of solutions of radionuclides are given in Table 12. All of these radionuclide solutions were poured into a 300-ml polyethylene wash bottle. The final volume of the mixture was adjusted to 200ml. The radionuclide mixtures, along with the stabilized isotopes (see Table 12), were then sprayed evenly onto the surface of the

Table 12. Preparations of Solutions of Radionuclides.

	Co-58	Cs-134	Sr-85	H-3
Original form:				
Activity	72.9 mCi/ml	54.4 mCi/ml	29.0 mCi/ml	0.25 mCi/ml
Solution	0.5 M HCl	0.5 M HCl	0.5 M HCl	Water
Dilute to 50 ml[1]:				
Activity	0.02 mCi/ml	0.004 mCi/ml	0.02 mCi/ml	0.0005 mCi/ml
Solution	0.5M HCl	0.5 M HCl	0.5M CHl	Water
Stable Isotope[1]:				
Amount	15 mg	4.5 mg	8.0 mg	
Crystal form	$Co(NO_3)_2.6H_2O$	CsCl	$Sr(NO_3)_2$	
Dilute to 25 ml[2]:				
Amount[3]	50λ	50λ	50λ	50λ
Solution	0.5 M HCl	0.5 M HCl	0.5 M HCl	Water
Sample[4],				
Amount[5]	1 ml	1 ml	1 ml	1 ml
Conc. HCl	4 ml	4 ml	4 ml	-
Water	395 ml	395 ml	395 ml	9 ml
Total Volume	400 ml	400 ml	400 ml	10 ml

Note:

[1] The mixture solution was composed of these two sections except 50λ ($1\lambda = 10^{-3}$ml) out of each 50 ml radionuclide solutions.

[2] This dilution was prepared for the Ge(Li) detector sample.

[3] The added amount was pipetted from 50-ml solution.

[4] "Sample" means the one that will be applied to a Ge(Li) detector for activity determination.

[5] The added amount was pipetted from a 25-ml solution.

solid waste (see Appendix II for spiking procedures). The initial gamma-ray exposure at 1 meter from the surfaces of the columns was about 0.8 mR/hr for the combined Co-58, Cs-134, and Sr-95 at the beginning of the experiment.

2.1.3 Sealants

A plastic pipe cement (Weld-on #717 for PVC, Industrial Polychemical Service, Gardena, CA) was found satisfactory for sealing all pipes and joints. However, for gas sealing inside the landfill units, a combination of Seelye electric plastic welder (Seelye Inc., Minneapolis, MN) and a layer of epoxy sealant (Devcon Corp., Danvers, MA) was found to be adequate.

2.1.4 Gas Collection Indicator Solution

An excess amount of Na_2SO_4 was added to distilled water with continuous stirring until it was fully saturated. Concentrated H_2SO_4, at 30 ml/ℓ, was then added to the solution. A few milligrams of methylene red was then dissolved in the finished solution. This mixture was used to prevent dissolution of the gases, e.g., CO_2, into solution during gas collection.

2.1.5 Scintillation Solution

Seven gm of PPO (2,5-Diphenyloxazole), 1.5 gm of bis-MSB (p-Bis(o-methylstyryl) benzene), and 500 ml of Triton N-101 were added to each liter of p-Xylene.

2.1.6 Standard Strontium Solution

Different standard strontium solutions ranging from 0.5 mg/ℓ to 20 mg/ℓ in concentration were prepared from a standard strontium solution having a concentration of 1000 mg/ℓ (supplied by Fisher Scientific Company).

2.1.7 Standard Cobalt Solution

The standard cobalt solutions were prepared by dissolving different

amounts of $CoCl_2 \cdot 6H_2O$ (Reagent Grade, J. T. Baker Chem. Co.) in distilled water. The final concentrations ranged from 0.5 ppm to 20 ppm.

2.1.8 Standard Cesium Solution

Different amounts of C_sCl (Certified Grade, Fisher Scientific Co.) were dissolved in distilled water to yield concentrations between 0.5 ppm and 20 ppm.

2.2 Methods

2.2.1 Analysis of Leachate

Leachate samples were characterized on the basis of both gross properties and specific analyses including organic matter and heavy metals. The organics were broadly classified on the basis of parameters such as particle size and molecular weight. This technique was based on the work by Chian et al. (7) using centifugation, millipore filtration, membrane ultrafiltration and gel permeation.

A Ge(Li) detector was used to determine the activity of Co-58, Cs-137, and Sr-85, whereas a liquid scintillator was used for H-3 determination. A Geiger counter was employed to monitor the movement of the radionuclides within the lysimeter system.

2.2.1.1 Gross Parameters

Leachate samples obtained from the experimental landfills were analyzed for five-day biochemical oxygen demand (BOD_5), total carbon (TC), total inorganic carbon (TIC), total organic carbon (TOC), chemical oxygen demand (COD), total suspended solids (TSS), volatile suspended solids (VSS), pH, oxidation-reduction potential (ORP), alkalinity, and conductivity. TC, TIC, and TOC were determined with a Beckman Model 915 Total Organic Carbon Analyzer (Fullerton, CA), pH and ORP were measured with a Fisher Model 144 pH Meter, conductivity was measured with a

Yellow Springs Instrument Co. Model 31 Conductivity Bridge (Yellow Spring, OH), and the remaining analyses were performed according to Standard Methods (57).

2.2.1.2 Classification of Organic Matter in Leachate

This methodology was based on the fractionation scheme for the separation of particulates and molecular weight distribution of soluble organics reported by DeWalle and Chian (12) (see Figure 5). Each of the separation processes are described as follows:

Centrifugation and Millipore Filters

The leachate was first centrifuged (Modal 61 Centrifuge, Chicago Surgical and Electrical, Chicago, IL) followed by filtration through a 0.45 μm membrane (Gelman, Ann Arbor, MI). This step will remove suspended solids and microorganisms.

Membrane Ultrafiltration and Gel Permeation

Concentration and separation of organics based on the molecular weight cut off of membranes was accomplished by dialyses, ultrafiltration (UF) or reverse osmosis (RO). Dialysis is based on diffusion in which relatively small solutes permeate the membrane under the driving force of a concentration gradient, while larger solutes are retained due to the sieving effect of the membrane (27). Ultrafiltration operates on the principle of sieving and separates from a solution those organics whose molecular dimensions are ten or more times larger than the solvent, whereas the solvent flows through a membrane under a pressure driving force (36). Reverse osmosis works on a principle similar to ultrafiltration except the solute transport occurs by diffusional mechanisms and an appreciable osmotic pressure builds up in reverse osmosis. The solvent transport is by viscous flow mechanisms as in ultrafultration (11).

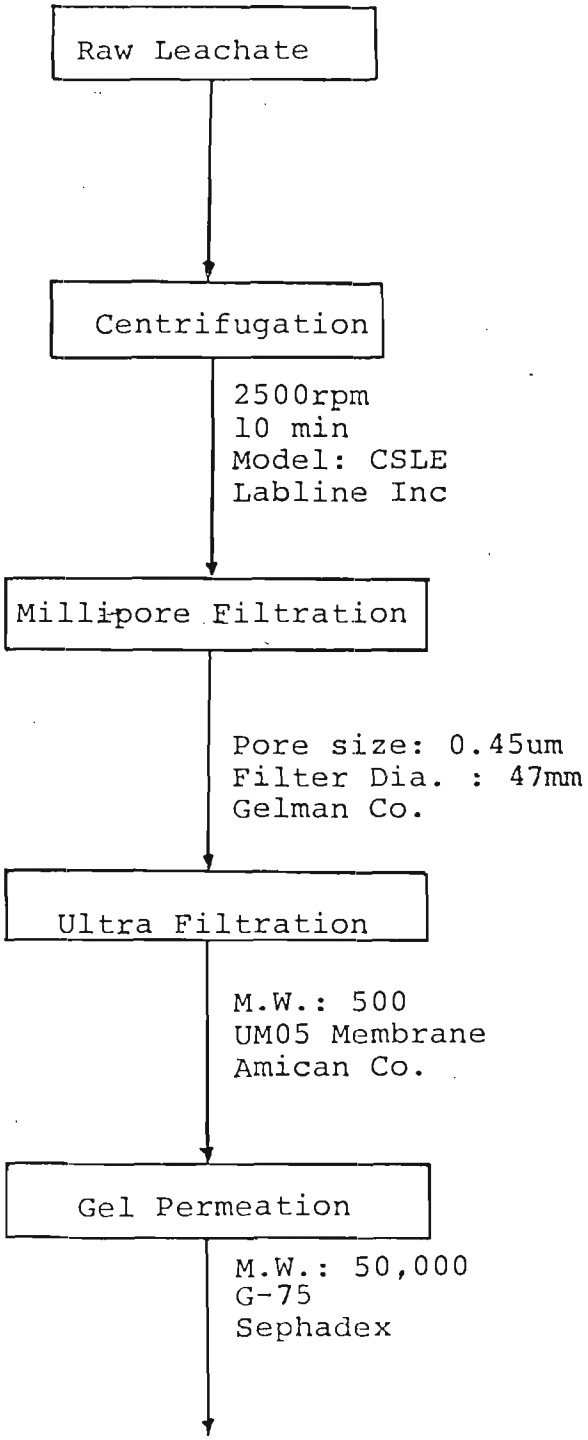


Figure 5. Separation Scheme of Organics in Leachate.

In a study of the comparison of membrane separation with other separation techniques, Chian et al. (11) reported that the combined use of membrane ultrafiltration followed by gel permeation of the UF retentate gave the best separation of organics present in leachate. A 500 MW cut off UF membrane (e.g., Amicon UM04, Bedord, MA) and a Sephadex G-75 column, (Pharmcia, Piscataway, NJ) were employed to separate the organic in different molecular weight fractions. Aanalysis of functional groups, such as carboxylic, phenolic, proteins and carbohydrates, were performed on the gel permeation fractions of UF retentates, e.g., the humic and the fulvic acid fractions, whereas that of the low-molecular-weight volatile fatty acids, on the UF permeate. The humic fraction refers to whatever organics are excluded by the G-75 resin (i.e., the high MW fraction), whereas the fulvic fraction refers to the remainder of the organics retained by the UF membrane (i.e., the lower MW fraction).

The molecular-weight distribution of the organic compounds in the UF retentate was examined by fractionation on Sephadex G-75 having exclusion limits of 1000 to 50,000. A 10mm inner diameter glass column was filled with swollen Sephadex. The sample volume injected was 0.5 ml. The eluant was collected in 10-ml fraction each. Water was used as the mobil phase. Since the volume in which the maximum concentration of the compound is directly proportional to the logarithm of the molecular weight of the compound, standardization of the Sephadex column was necessary to estimate the apparent molecular weight. Blue dextran (with MW ~ 2,000,000), ribonuclease A (with MW ~ 13,700), ovalbumin (with MW ~ 43,000), chymotrypsinogen A (with MW ~ 25,000) and albumin (with MW ~ 67,000) were used to calibrate the column and determine the volume in which the maximum concentration of each molecular-weight fraction eluted. Based on this calibration, fractions

were collected as follows:

1st cut: High MW fraction, with MW > 50,000

2nd cut: Medium MW fraction, with 1000 < MW < 50,000

3rd cut: Low MW fraction, with MW < 1000

2.2.1.3 Volatile Fatty Acids

A Hewlett-Packard Model 5710 A (Avondale, PA) gas chromotograph equipped with a flame ionization detector was used to measure the concentration of the volatile fatty acids. A 2 meters long by 2mm I.D. packed column (3% carbowax 20m, 1.5% phosphoric acid on carbopack B #60 - #80 mesh size) was used to separate acetic- propionic, butyric, iso-butyric, and valeric acids. Filtered weekly samples were analyzed immediately after collection.

The gas chromatograph (GC) conditions were as follows:

Injection Temp	250°C
Detector temp	FID @ 250°C
Oven	Temperature Programming initial temp- 105°C for 2 min, final temp - 140°C for 30 min, rate 4°C/min
Carrier Gas	N_2 @ 40 ml/min

2.2.1.4 Heavy Metals

The metals analyzed in this study included: iron, magnesium, manganese, potassium, sodium, calcium, and nonradioactive strontium, cobalt, and cesium. The concentration of each metal was determined by using an atomic absorption spectrophotometer. Two different atomic absorption spectrophotometers were employed, i.e., Perkin Elmer Model 303 and Model 703 (Norwalk, CT). The former was used for metal concentration in the mg/ℓ range, and the latter for metal concentrations in the µg/ℓ range.

Leachate samples were digested prior to analysis with the spectrophotometer. The procedures for sample digestion were as follows: To 100 ml of sample that was placed in 250-ml beaker, 5 ml of 1:1 diluted ultrapure HNO_3 (Ultrex) were added covered with a watch glass and evaporated on a hot plate at 95°C (without bubble forming) to near dryness. After the sample was cooled, 4 ml of concentrated Ultrex were added, and again evaporated to near dryness. After cooling, 1 ml of 1:1 Ultrex and 3 ml of 30% H_2O_2 were added. The beaker was then heated until the effervesence from the addition of H_2O_2 subsided. After cooling, the addition of 30% H_2O_2 in 1 ml aliquots was repeated until the effervescence was minimal or the general sample appearance was unchanged. No more than a total of 10 ml of 30% H_2O_2 was added. After cooling, 1 ml of 1:1 Ultrex was again added. The sample was then refluxed for 10 minutes. After cooling, the sample was diluted to 100 ml and stored in a polyethylene container (63).

Digestion blanks of doubly distilled water were prepared for each digestion scheme presented above. They were used as a correction for possible metal introduction due to the digestion procedures.

Sample preservations were employed in accordance with the EPA recommended procedures for metals (64). Five ml of concentrated Ultrex were added to each liter of sample and the acidified samples were then stored in a refrigerator at 4°C.

2.2.1.5 Radioactivity

400 ml of filtered leachate sample was measured with a Ge(Li) detector for the determination of Co, Cs, and Sr activities. The instruments include: Ge(Li) detector, preamplifier, high voltage power supply, spectroscopy, 4096-channel analyzer, printer, and calculator plotter. The detector had

been calibrated with an NBS source for counting efficiency as a function of gamma ray energy.

To determine H-3, four ml of the filtered leachate sample was distilled. Two ml of the condensed water was then pipetted into a 25 ml vial and mixed with 18 ml of scintillation solution. A standard solution (prepared with a known amount of H-3) and a background solution (distilled water) were employed along with the sample solution while using a liquid scintillator (Beckman LS-233) for H-3 activity detection (3).

2.2.2 Analysis of Gases

The gas generated from the landfill units was collected and analyzed for the quantity, composition, and radionuclides (which might appear in the form as H_2, H_2O, and CH_4). The percentages of CH_4, CO_2, N_2, and O_2 in the gas were analyzed by employing a gas partitioner (Fisher Scientific Co. Model 25V) along with a thermal stabilizer (Fisher Scientific Co. Model 27), where helium was used as the carrier gas. For H_2, the same instruments were used, but argon was used as the carrier gas.

Although H-3 exists mostly in the form of water in the liquid phase, there is a possibility that it may be present as a component in the methane and water vapor phases. The determination of H-3 in the gases generated from the units was therefore necessary for this study. The procedures for H-3 determination in the gas phase are outlined in the following:

1. The gas sample was first passed through a Na_2SO_4 (anhydrous) column. The water vapor was absorbed by the column, while CH_4 and other gases (e.g., CO_2) passed through.

2. The effluent gas stream passing through the Na_2SO_4 column was mixed with air and burned completely in a catalytic TOC conversion furnace

(Lindberg Co. Type 55035). During this process, CH_4 was oxidized to final products H_2O and CO_2.

3. The gas stream then flowed through a second Na_2SO_4 column where the H_2O was adsorbed by the column and the CO_2 passed through.

4. The effluent gas stream from the second Na_2SO_4 column was introduced into a NaOH solution. As the gas stream passed the NaOH solution, the CO_2 gas was absorbed.

5. The Na_2SO_4 was collected and stored in a self-sealed container. The water content in the Na_2SO_4 crystals was removed by heating and condensation. The collected water was then applied to a liquid scintillator for H-3 determination. A schematic diagram of this method is shown in Figure 6.

2.3 Experimental Techniques

Two identical polyethylene containers with conical bottoms were employed as experimental columns in this study. One container was operated as the control unit without leachate recycle, the other as the test unit with leachate recycle. A schematic diagram of the system is shown in Figure 7. A more detailed diagram of the test unit, i.e., the recycle unit, is shown in Figure 8. Each container has an inner diameter of 57 cm. The height of the cylindrical section is 74 cm, and the conical section is 15 cm. The overall volume is 208 l (55 gallons). Due to the limited, yet rather crucial amount of the radioactive isotopes applied to the landfill units, the pebble-shaped media should be made of nonporous material (in this study, marble was used), so that the adsorption of the radioactive isotopes on them can be minimized. The conical section was then filled with marbles to prevent clogging and facilitate collection of leachate by the under drain pipe. A perforated polyethylene pipe was installed at the

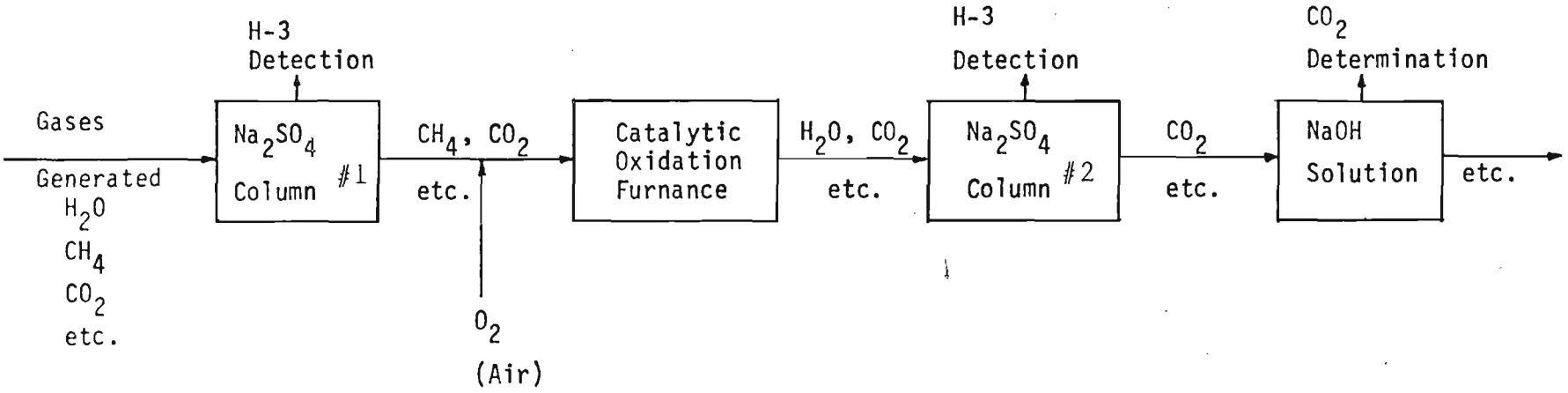


Figure 6. Schematic Diagram of H-3 Determination Procedures

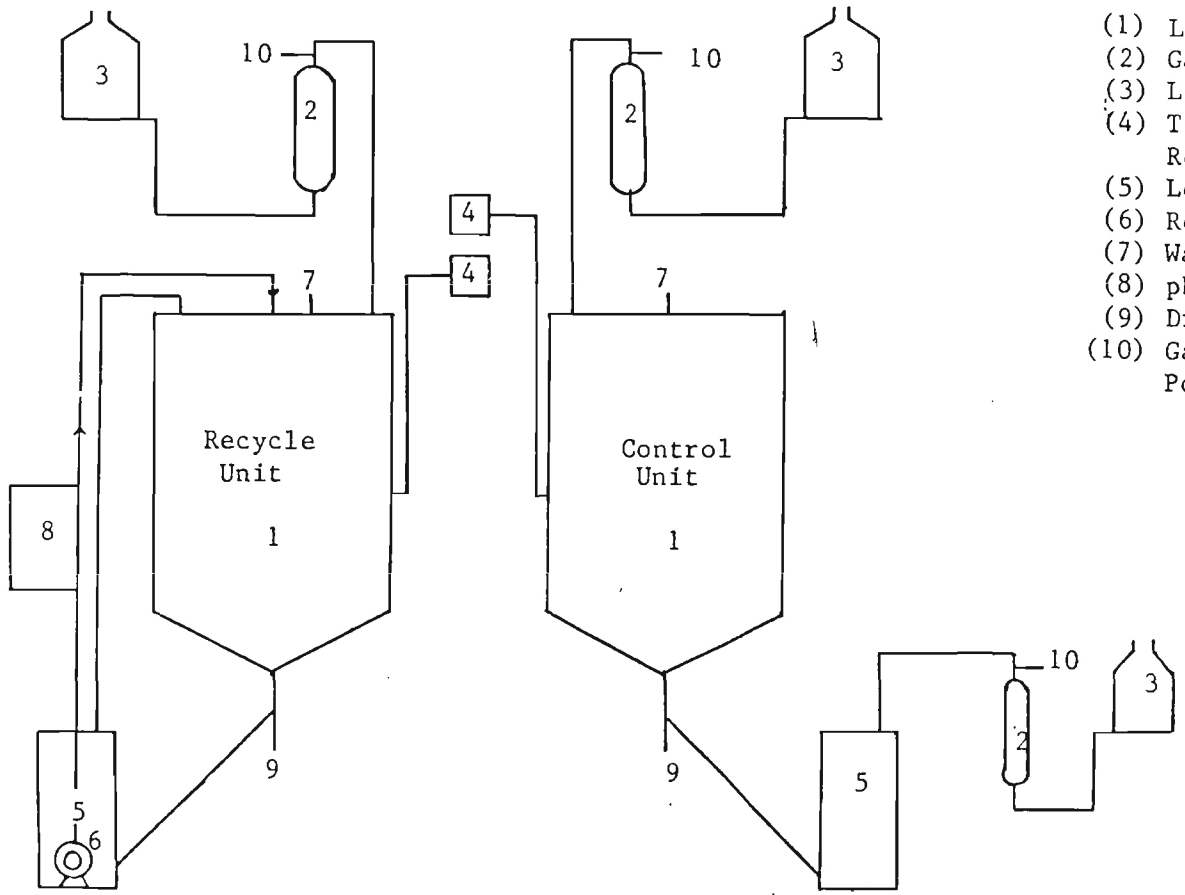


Figure 7. Schematic Diagram of The Laboratory Simulated Landfill System

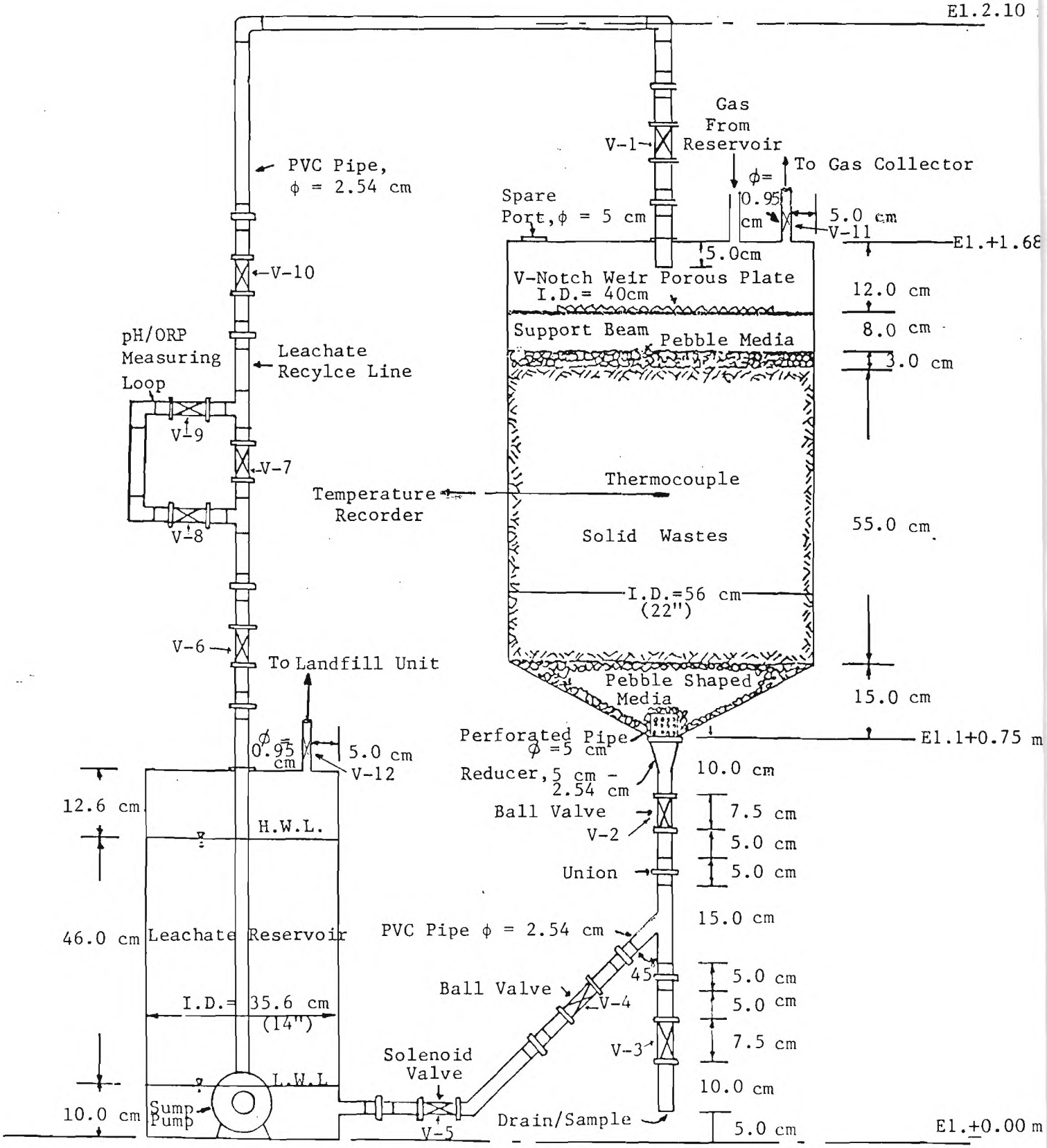


Figure 8. Detailed Drawing of the Lab-Scale Landfill Unit with Leachate Recycle

bottom of the container for the purpose of leachate collection.

The leachate recycle pump was operated on a daily basis. A solenoid valve, installed on the leachate collecting pipe, was shut off during each pumping period. An event recorder was connected to the solenoid valve, allowing an estimate of the amount of leachage recycled each day.

A thermocouple was located in the center of the compacted solid waste. To protect against corrosion the thermocouple was encased in a heat-shrinkable tube coated with a sealing material. A recorder was used to monitor temperature inside the landfill units.

For safety, the entire systems were placed on metal trays to collect any possible leaks or spills. The working area around the landfill units was isolated in a separate room with a locked door. Radiation warning signs were placed to prevent entrance of unauthorized personnel. The initial gamma-ray exposure at 30 cm from the surface of the landfill units has been estimated as approximately 1 mrem/hr for Co-58, Cs-134, and Sr-85, respectively, at the beginning of the experiment. A gamma-ray survey meter was kept on the premises.

A pH meter, equipped with an oxidation-reduction potential (ORP) measurement system, was used to determine the pH and ORP values of the leachate generated. The lead of the detecting probe was long enough so that daily readings could be taken at a remote distance to minimize personnel exposure to the radiation.

The Photographs of the lab-scale landfill systems are given in Plates 1-5.

Plate 1: The control unit without leachate recirculation is shown in Plate 1. It consists of a 208-liter tank, a 57-ℓ leachate reservoir, two gas collecting burets and leveling bottles, and

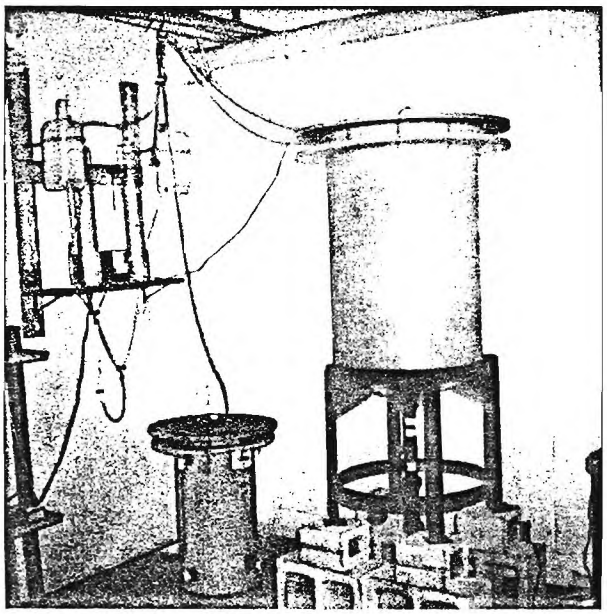
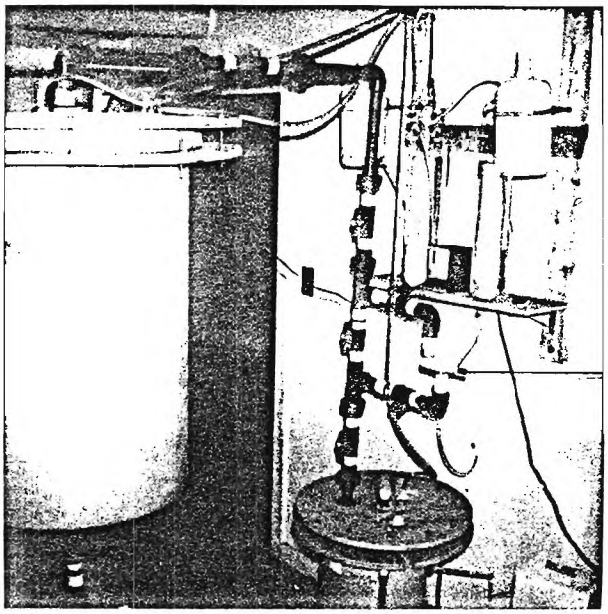


Plate 1



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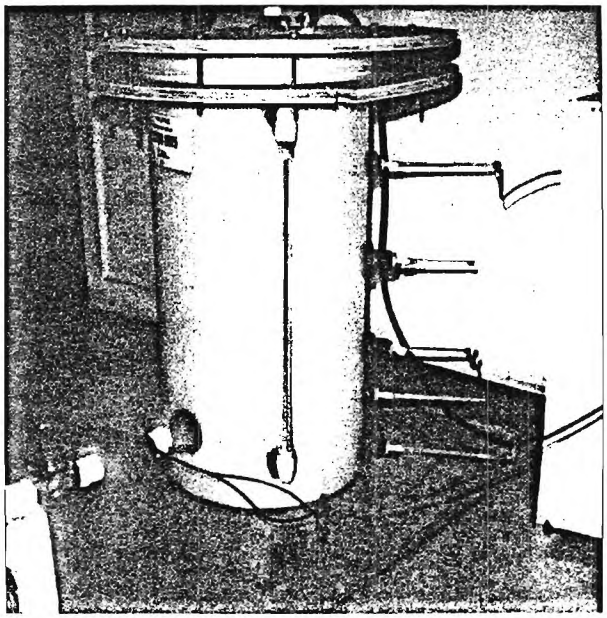

Plate 3

a temperature recorder. The entire unit is placed inside of an aluminum pan. The label on the tanks is marked with "Radioactive Materials".

Plate 2: The recycle unit is shown in Plate 2. The unit consists of the identical equipment items as the control unit with the exception that a recycle system is attached. The recycle system consists of a recirculation pipe line, a submerged sump pump (in the leachate reservoir), four level control probes, a pH/ORP loop, and a solenoid valve.

Plate 3: A close-up view of the leachate reservoir of the recycle is shown in Plate 3. There are four liquid level control probes mounted on the side of the reservoir, which allows control of the recycle liquid levels ranging from 10 to 40 liters. A Plexiglas side-arm is installed outside the unit to indicate the liquid level in the reservoir. The solenoid valve (attached to wires) is also shown in this picture.

Plate 4: The pH/ORP loop, the pH/ORP probe, and the pH/ORP meter are shown in this plate. The liquid level controller is located at the lower deck of the stand.

Plate 5: A close-up of the gas collecting buret and leveling bottles. The temperature recorder is located between the two burets.

Hydraulic and the pneumatic tests were conducted to locate possible leaks of liquid and gas from the lysimeters. The procedures for each test are detailed in Appendix I. Filling and spiking methods for the landfill units are detailed in Appendix II. Operating procedures for the system are detailed in Appendix III.

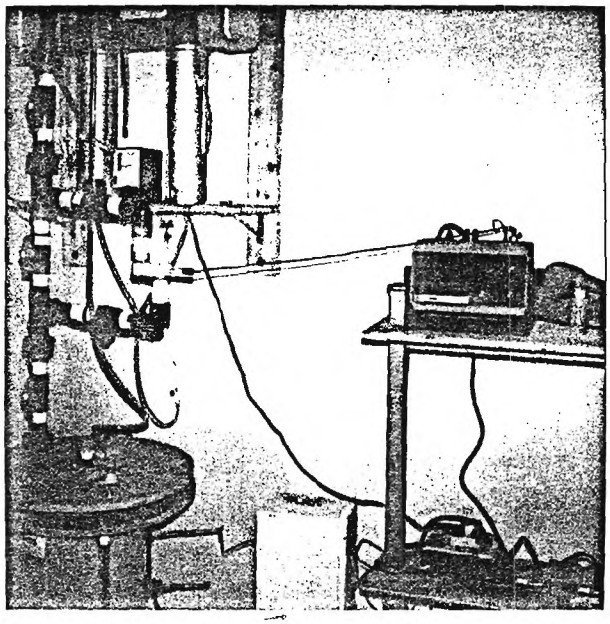

Plate 4

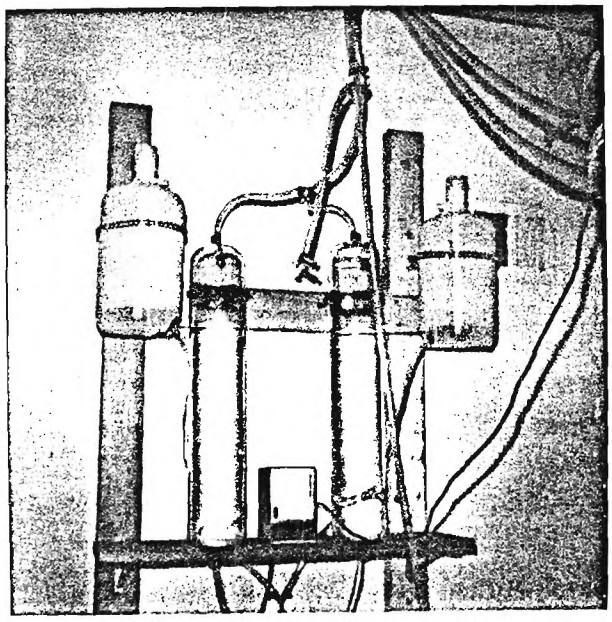

Plate 5

After operation, the entire system will be shut down. Once the destruction and packing processes have been completed, all the liquid and solid materials will be further treated and disposed of as regular low-level radioactive wastes.

III. RESULTS AND DISCUSSION

Results of the analyses performed on the leachate and gases generated and collected from the two simulated landfill units are presented in the following sections. Data relating to environmental conditions which existed during the test period are also presented. The time scale used in this presentation is time since leachate production began (May 18, 1981). Although research is continuing, this report covers the period to October 19, 1981 (Day 154 since leachate production began).

3.1 Lab-Scale Simulation of Landfill

The work involved with the design of the simualted landfill units was initiated at the end of 1980, and the final sealing of the landfill units was completed in May 1981. During this period, revision of the preliminary design, procurement of equipment, start-up procedures, and many related efforts had been conducted. A detailed description of the lab-scale landfill units is presented in the previous section 2.3.

3.2 Solid Waste Analysis

The solid waste used for this study had a moisture content of 22.5% and a volatile solid content of 77%. The carbon, hydrogen, and nitrogen contents were determined to be 64.4%, 6.1%, and 3.6%, respectively. Each of the two 208-liter (55-gal) containers was filled with 43 kg (dry weight) of shredded solid waste. The final volume of the solid waste after compaction was about 170 liters, and the density of solid waste was 250 kg/m^3 (dry weight) before adding water to attain the field capacity.

3.3 Environmental Conditions

The temperature inside the landfill units is shown in Figure 9, it varied with the daily fluctuation of ambient temperatures. For a mesophilic anaerobic system, the optimum temperature is around 33°C - 35°C. The room temperature was raised eventually to that range beginning in early July 1981 (48 days since leachate production began).

Fifty-three liters of distilled and deionized water was added to each container to attain the field capacity. Twelve liters of water was present in the refuse due to its moisture content. After the water was added to the container, it was then collected from the under drain pipe and pumped/poured back to the container daily for a period of one week.

The amount of distilled and deionized water added to each landfill unit as simulated precipitation was one liter per week. The one liter per week rate was derived from the observations of rainfall rates reported by Pohland (47).

Collections of the gas and leachate were initiated on the eighth day after solid waste placement. Results of analysis of leachate and gas samples generated from the control and the recycle units are presented and discussed as follows:

3.4 Leachate Analysis

The leachate samples were collected on a weekly basis. Approximately 800 ml of leachate was withdrawn weekly from each of the landfill units for analysis. After determinations of the gross parameters, organic matter, and radioactivities, a remainder of approximately 400 ml of the sample was poured back to the landfill units. The contaminated sample, approximately 400 ml, was stored in a bottle labelled with "Radioactive Materials." The amount of leachate recycled in the recycle unit initially was 19 ℓ/dav and subsequently increased to 38 ℓ/dav.

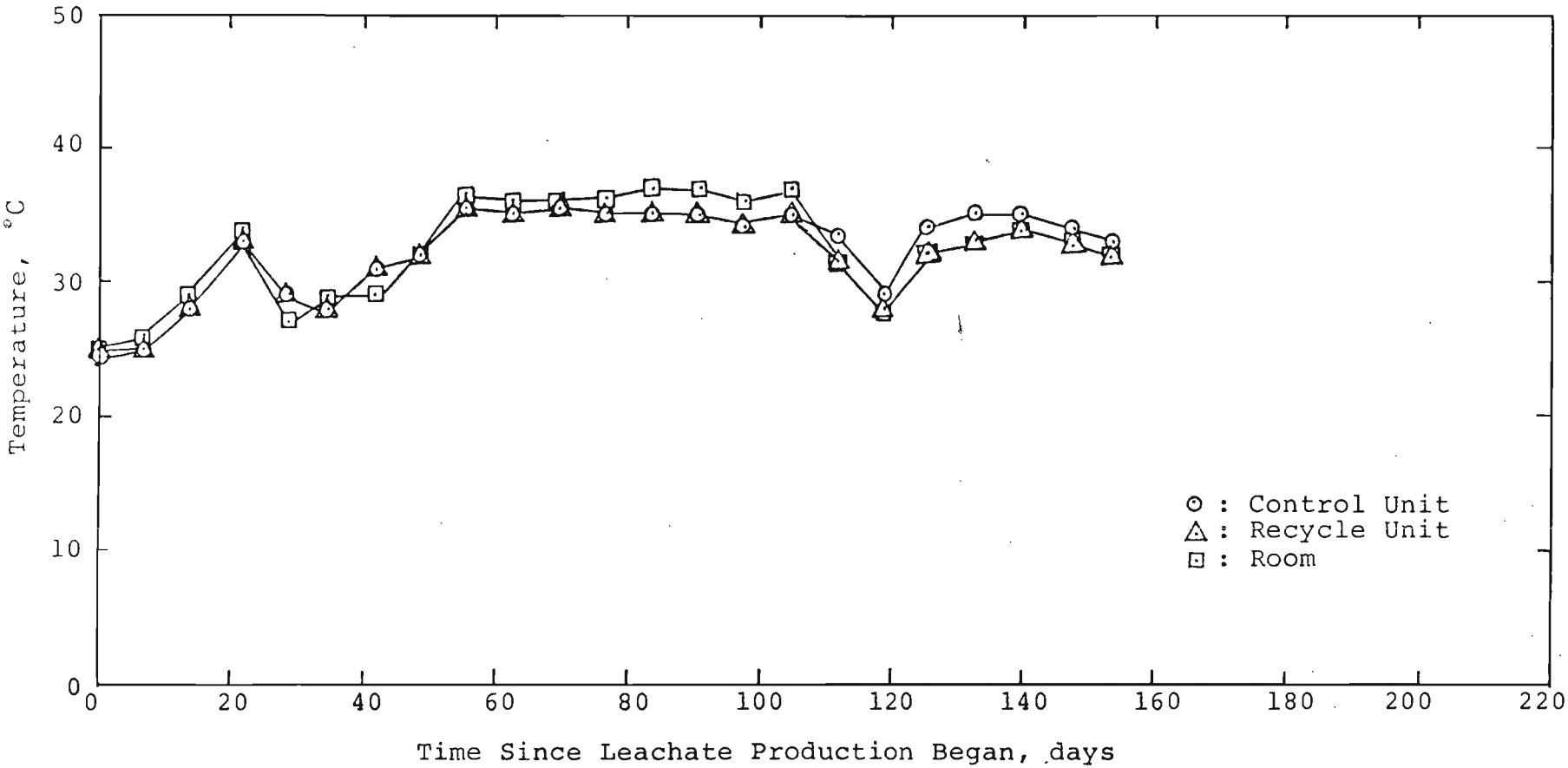

Figure 9. Temperature in Leachate

The values of COD, BOD_5, TOC, and ratios of BOD_5/COD, and COD/TOC are shown in Figures 10, 11, 12, 13, and 14, respectively. In dealing with anaerobic biological processes as exist within landfills, levels of COD, BOD_5, and TOC are most important parameters. Concentrations of these parameters tend to rise to a maximum and then decrease gradually as the anaerobic processes proceed and the landfill becomes stabilized. Figures 10, 11, and 12 show the same trend for both the control and the recycle units. The values of COD, BOD_5, and TOC decrease significantly during the same period when the TSS and pH increase sharply and the ORP decreases rapidly below -200 mV. As can be seen in the later section of this report (Figure 17), the total volatile fatty acids also decrease rapidly during this period. The lower value of total volatile fatty acids as well as that of the BOD_5/COD ratio (Figure 13)also confirm that the leachate of both units are being anaerobically stabilized.

The initial values of COD in leachate from the control and the recycle units were 18,600 mg/ℓ and 22,800 mg/ℓ , respectively; ultimate values were 1,900 mg/ℓ, and 3,800 mg/ℓ, respectively. Initial values of BOD_5 in leachate from the control and the recycle units were 9,300 mg/ℓ and 11,500 mg/ℓ, respectively. The initial values of TOC in leachate from the control and the recycle units were 7,000 mg/ℓ and 7,800 mg/ℓ, respectively; ultimate values were 930 mg/ℓ and 1,430 mg/ℓ, respectively.

Although a rapid decline in leachate pollutant concentrations (as expressed in terms of COD, BOD_5 and TOC) were observed for both landfill units, somehow the removal mechanisms are different. As for the recycle unit, daily recycle of leachate provided a continuing exposure of the internal biological populations to nutrients contained in the leachate and thereby enhanced overall conversion of those constituents as well as those in the solid waste and transfered to the leachate during passage. Whence

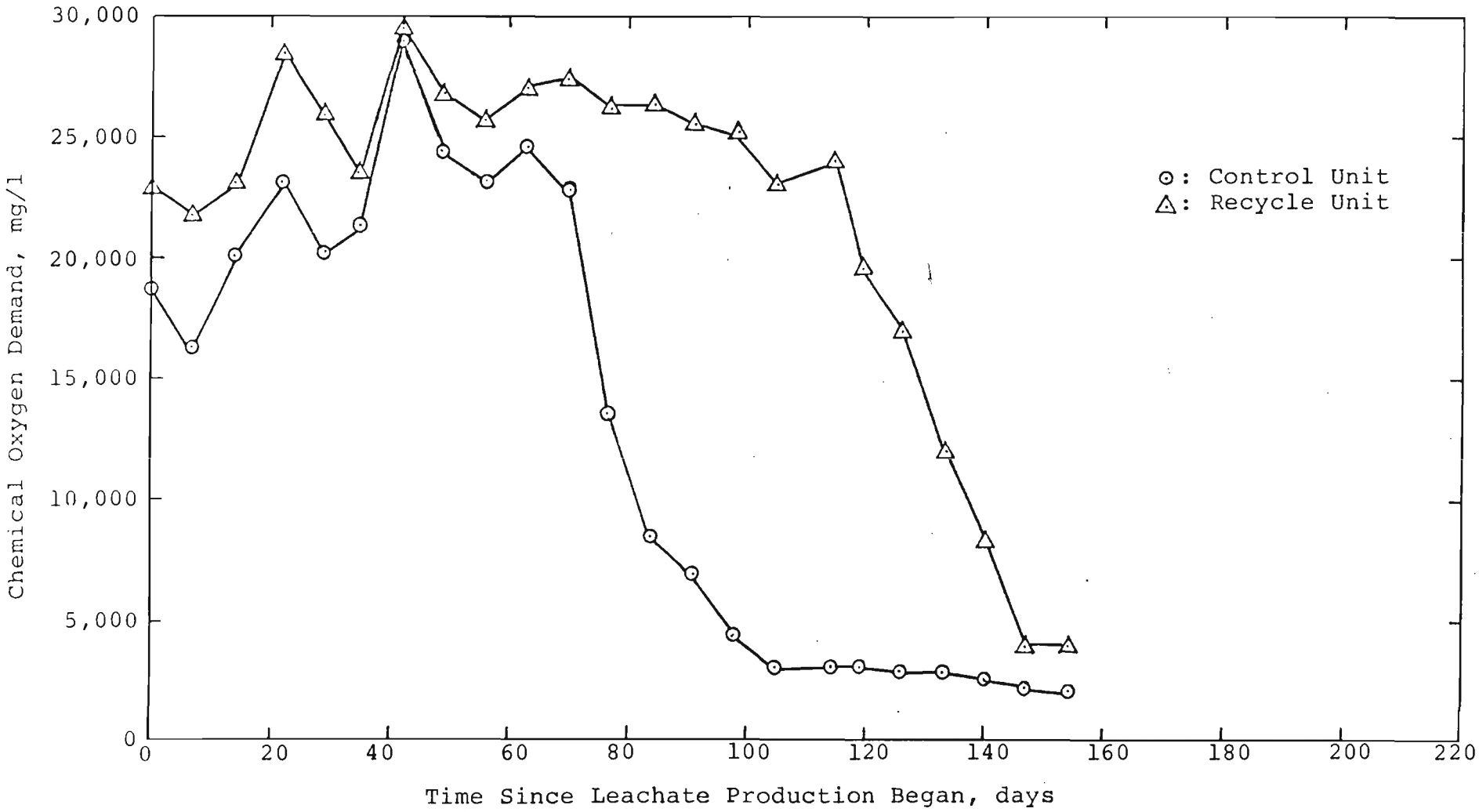

Figure 10. Chemical Oxygen Demand of Leachate

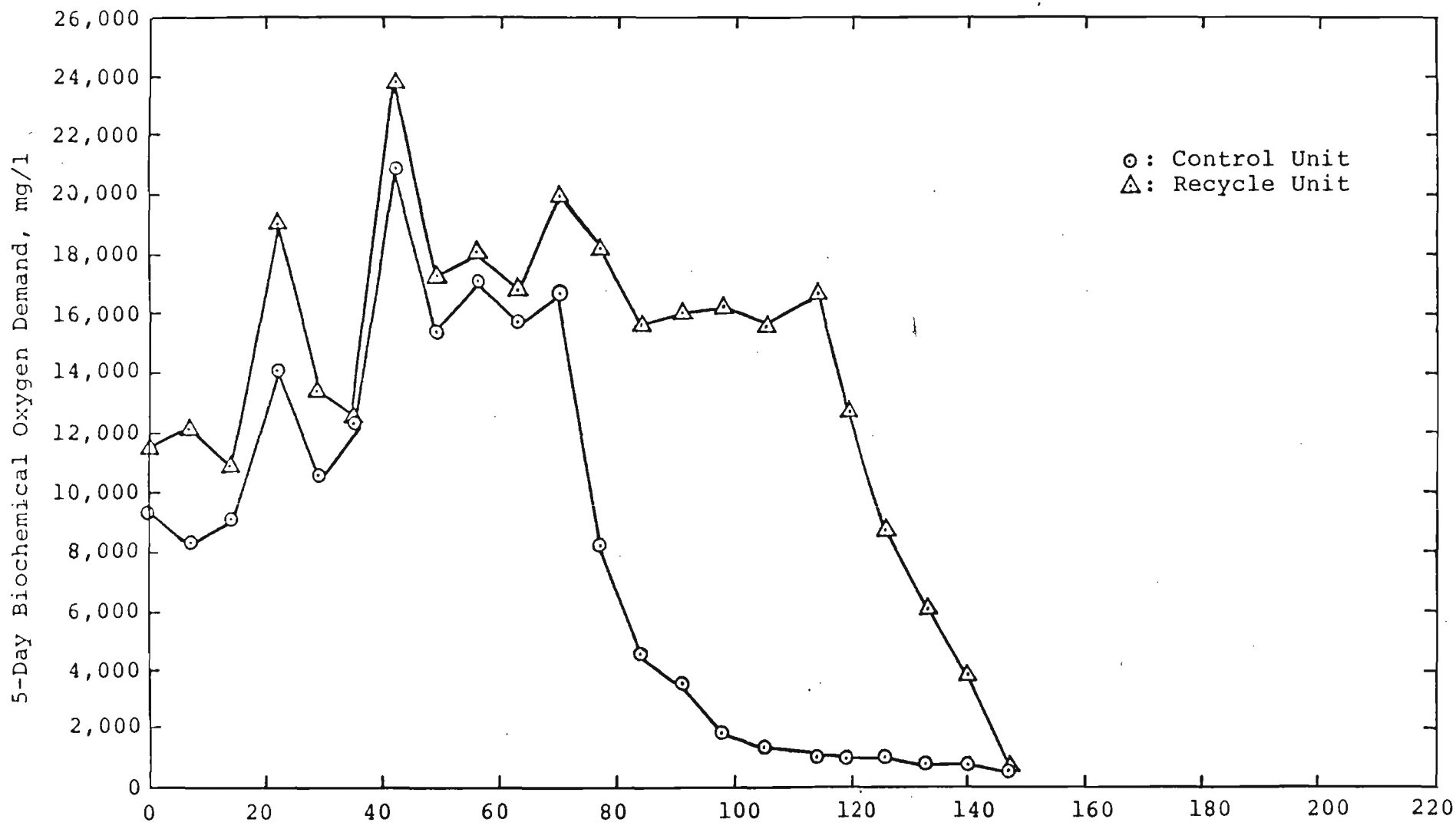

Figure 11. 5-Day Biochemical Oxygen Demand of Leachate

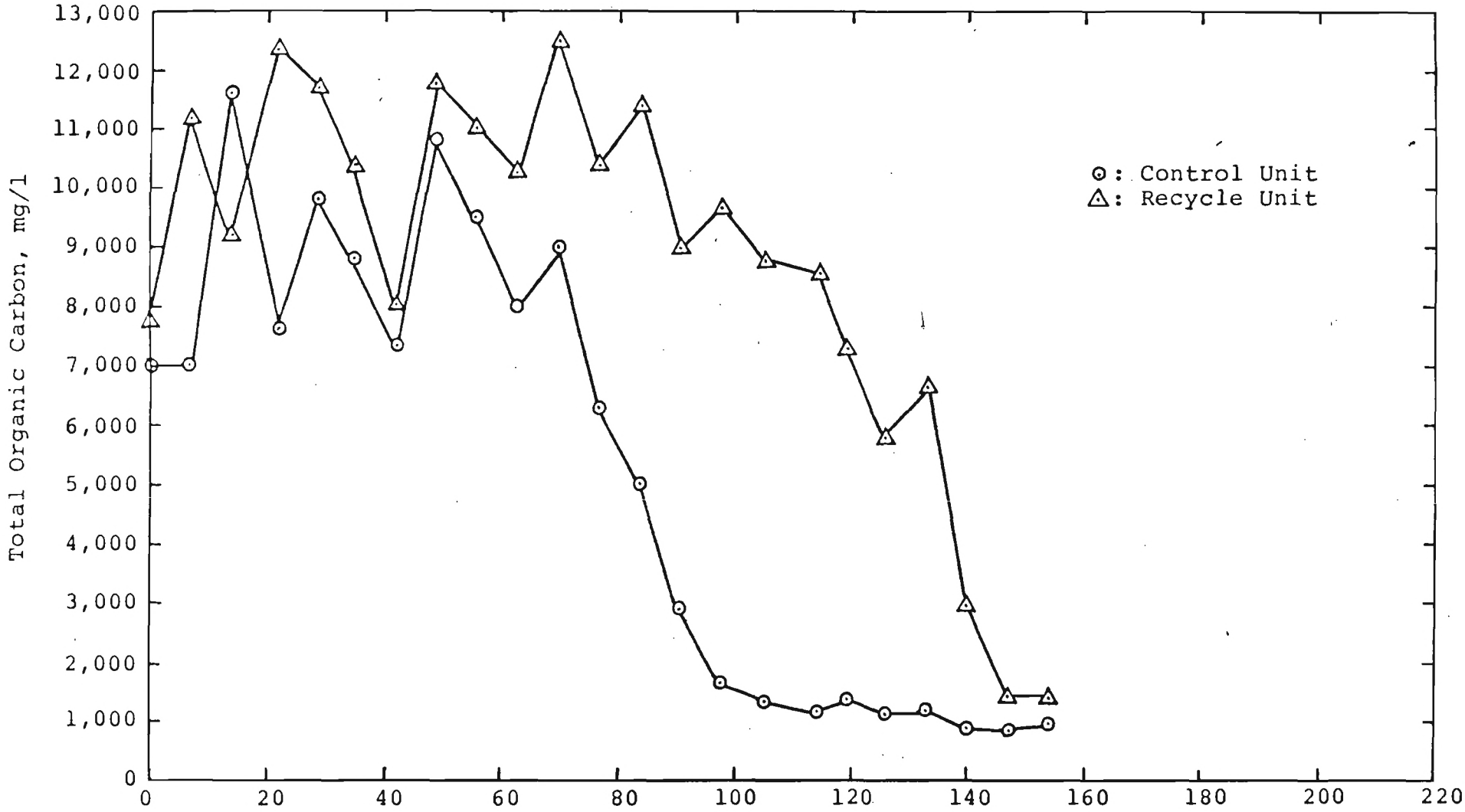

Figure 12. Total Organic Carbon of Leachate

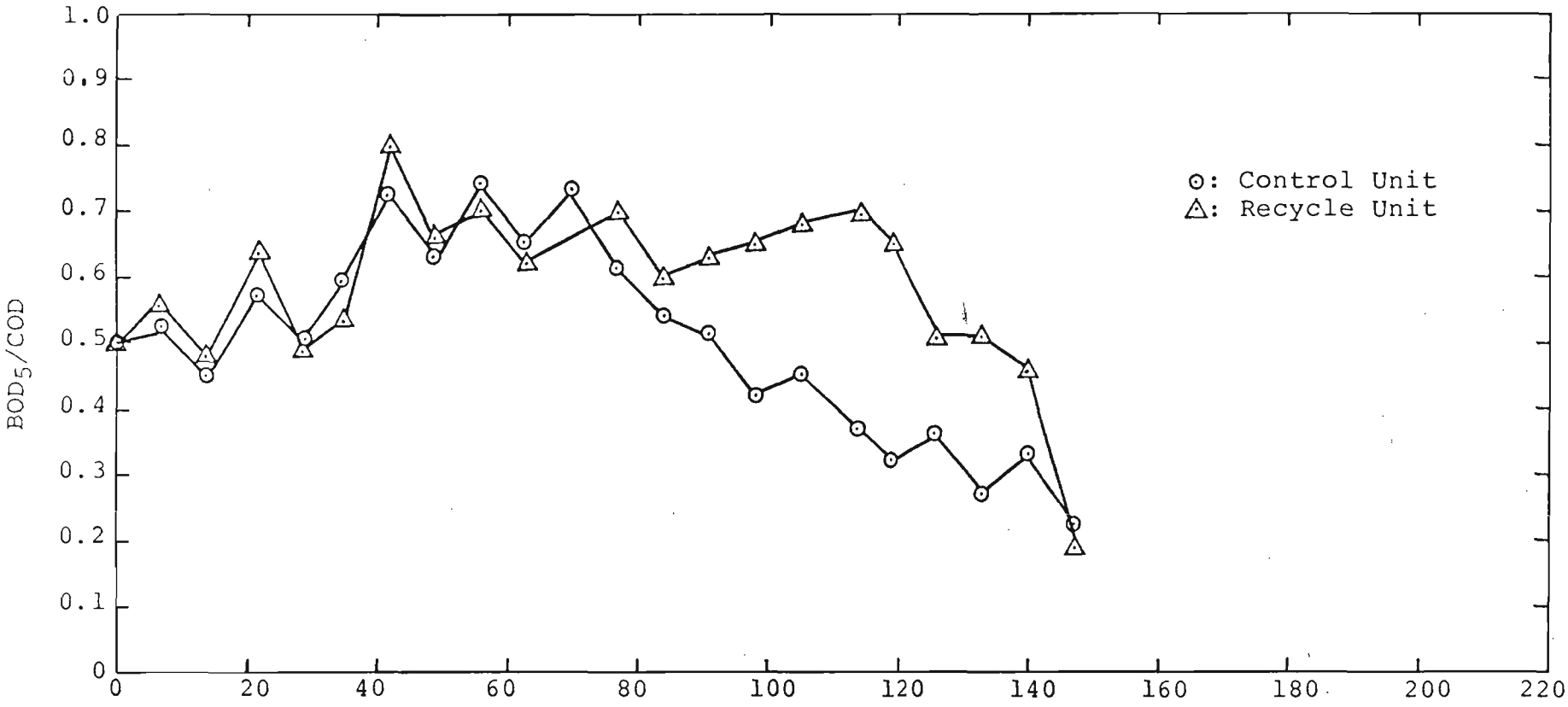

Figure 13. Ratio of BOD_5/COD of Leachate

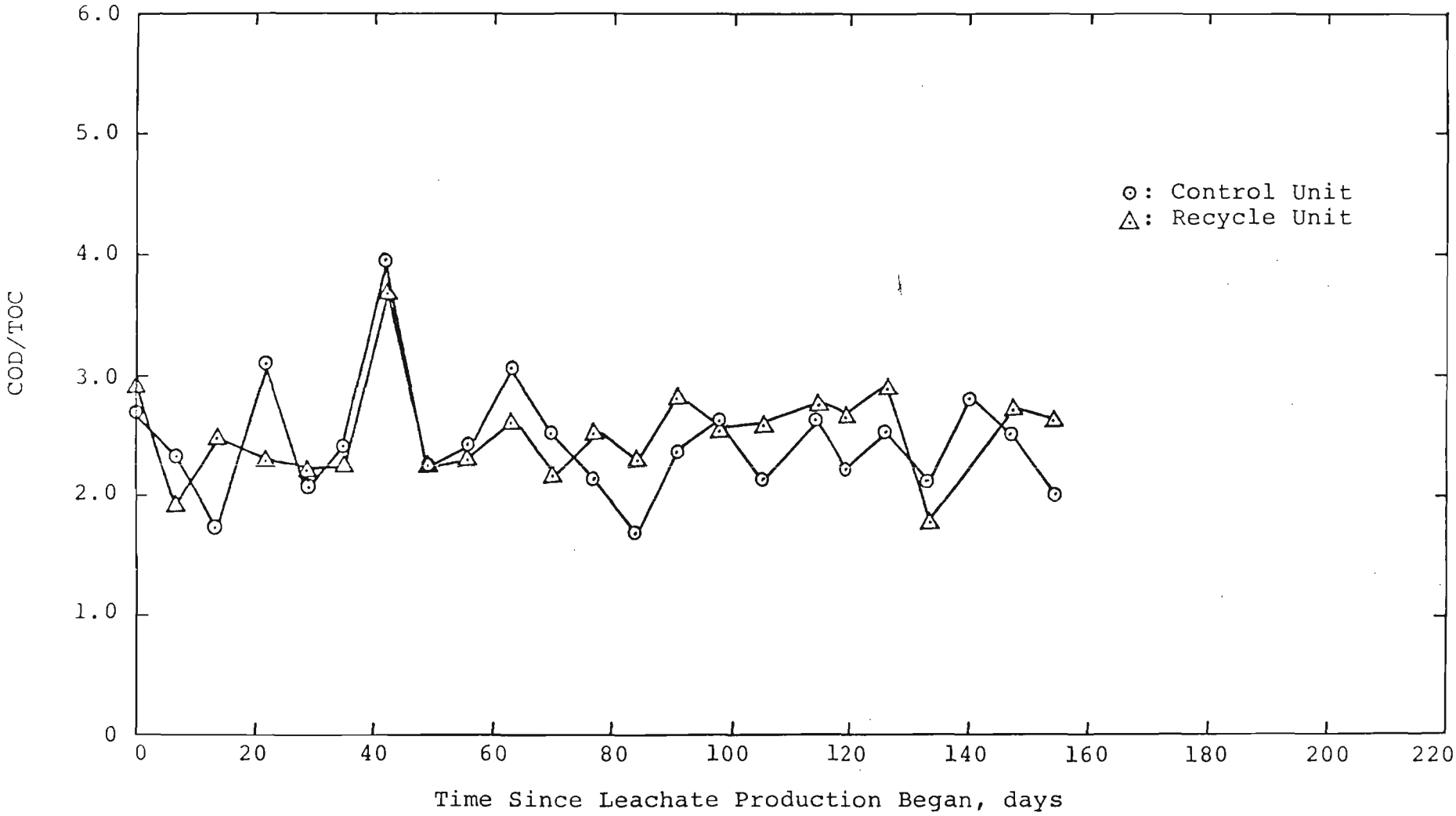

Figure 14. Ratio of COD/TOC of Leachate

daily recycle was considered primarily indicative of an initial acceleration of biological stabilization of the more readily available organic materials contained within the recycle unit. Whereas for the control unit, the organic materials within the solid waste had been washed out by the single-pass weekly added water, which made less available nutrients for the biological populations. As a result by this single-pass system, a lower value of organic pollutants were measured in the leachate of the control unit. Consequently, a lower percentage of organic pollutants removal was resulted, and the potential of leachate pollution was still unsolved.

For an anaerobic processes, the final products are CH_4 and CO_2, so that the amount of gas produced reflects the completeness of removal of organic pollutants. The cumulative gas produced amount for both landfill units (See Figure 30) confirmed the above statement.

As can be seen in Figure 13, the ratio of BOD_5/COD for leachate from the control unit increased initially from 0.5 to 0.74, and then decreases gradually to 0.22; that from the recycle unit increased from 0.5 to 0.8 and then decreased to 0.19. The value of COD/TOC for the control unit increased from 2.7 to 3.9, and then decreased to 2.0; that for the recycle unit increased from 2.9 to 3.7 and then decreased to 2.7 (See Figure 14). Chian and DeWalle (9) have shown that as the landfills reached their final stages of stabilization, BOD_5/COD nad COD/TOC decrease to values of less than 0.05 and 1.3, respectively. Based on the values of these two ratios obtained in this study, i.e., BOD_5/COD and COD/TOC, the simulated landfills were approaching the final stages of stabilization.

Further interpretations of the observed changes in pollutional characteristics of the leachate can be based on the production of intermediates such as the volatile fatty acids. Analysis of volatile fatty

acids included acetic acid, propionic acid, iso-butyric acid, butyric acid, and valeric acid. The GC was calibrated prior to sample analysis by preparing standard solutions of the fatty acids at concentrations of 1000, 500, 250, 100 and 50 mg/ℓ. The linear range and minimum detectable limits (MDL) were established based on this calibration curve. Repetitive injections at each level (at least 4 injections) were made to establish the reproductivity of the GC response. Based on the data, the MDL was found to be 50 ng/μℓ, or 50 ppm. Leachate samples were diluted to concentrations which lay within the linear range. Concentrations of each acid in the leachate samples from the control and the recycle units are shown in Figure 15 and 16, respectively. The concentrations of total volatile fatty acids as acetic acid are shown in Figure 17. All acids increased from original concentrations to a maximum between Days 40 to 50 (early July 1981) and then started decreasing (Figures 15, 16, and 17). Acetic acid was by far the most predominant species in the leachate samples. Acetic acid concentrations of the control unit increased from 2,700 mg/ℓ to maximum of 13,800 mg/ℓ, and then decreased to zero (or below the MDL); those of the recycle unit increased from 3,700 mg/ℓ to 8,500 mg/ℓ, and then also decreased to zero.

Inspection of Figure 15 indicates that acetic acid drastically decreased in the leachate from the control unit which corresponds to a rapid rise in pH (Figure 18) as well as an organism population shift (i.e., from the acid formers to the methane formers). In anaerobic systems, once the readily available organics have been transformed into volatile acids by the acid formers, methane formers will begin to develop, especially when the available food, i.e., acetic acid, etc. is abundant and environmental conditions are favorable. The methane former can convert acetic acid very rapidly to CH_4 and CO_2 which explains the sharp decrease of acetic acid

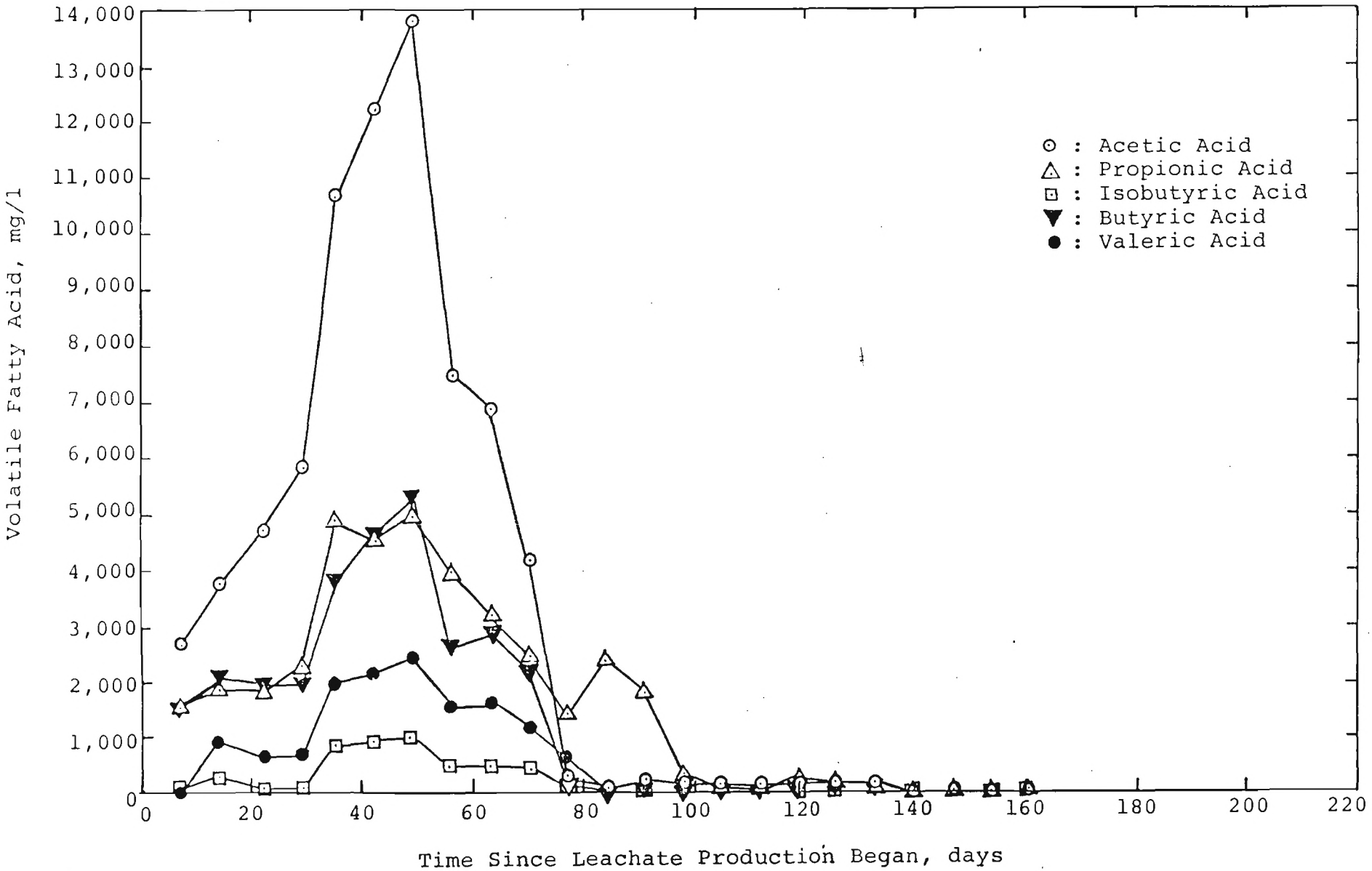

Figure 15. Volatile Fatty Acids of Leachate from the Control Unit

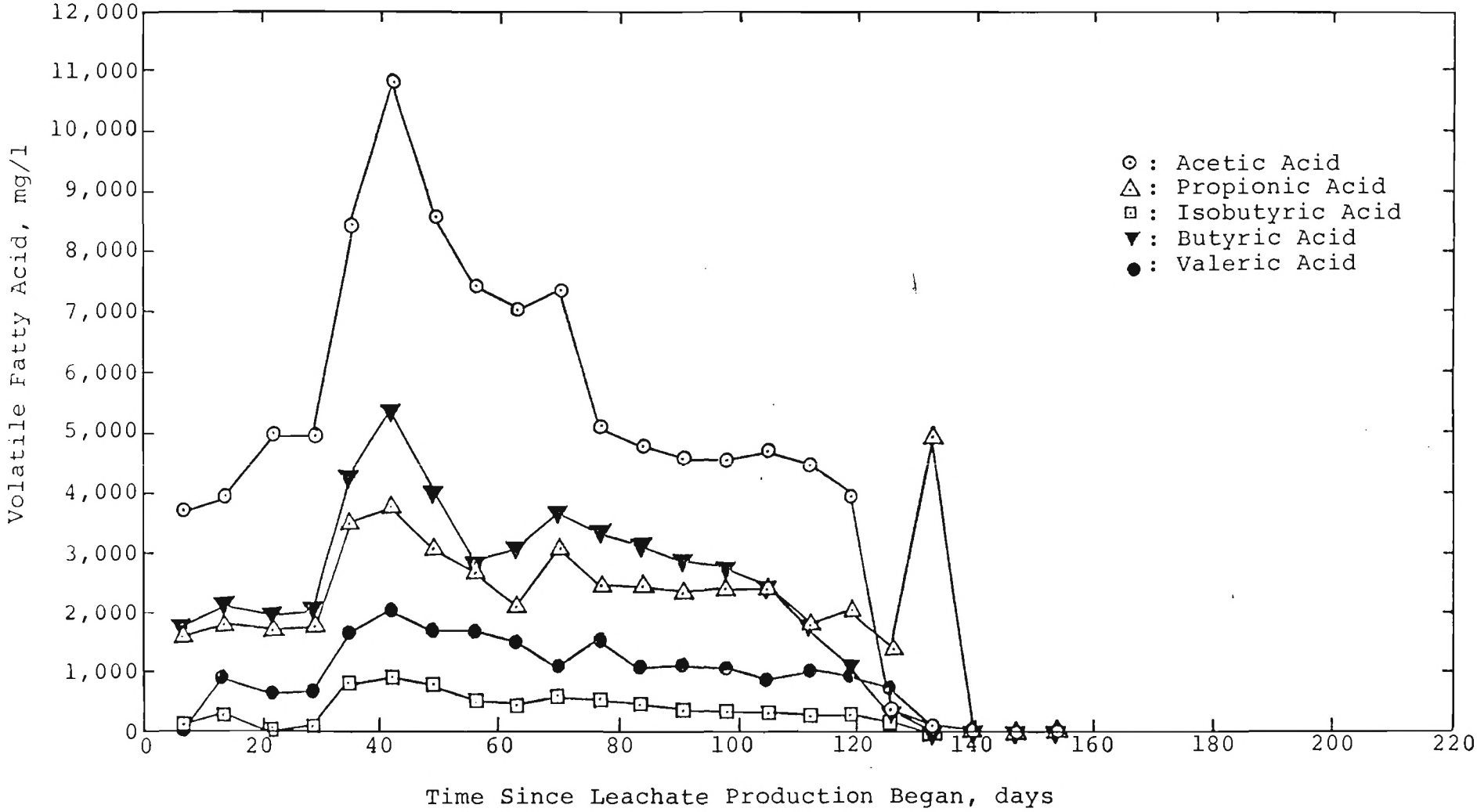

Figure 16. Volatile Fatty Acids of Leachate from the Recycle Unit

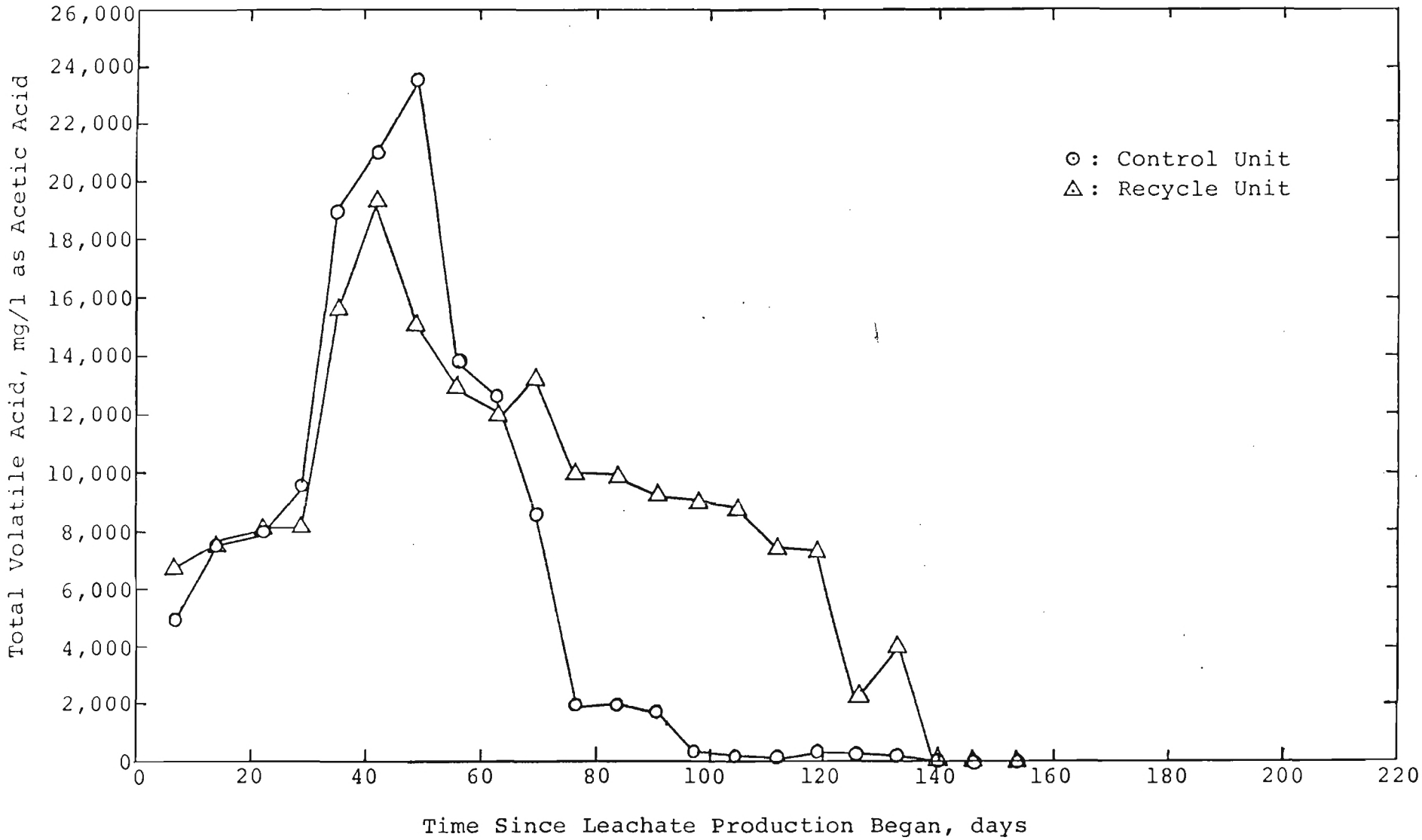

Figure 17. Total Volatile Acids of Leachate

concentrations and the increase in gas production (Figure 30). It also explains, (besides washout effects), why the pH in the control unit increased from 5.3 to 6.1 and then 7.0 within three week's time (see Figure 18) after the acetic acid peaked out; corresponding methane concentrations increased rapidly from 19% to 29% and 45% (see Figure 31). This increase in methane production supports the aforementioned shift of populations of microorganisms from acid formers to methane formers.

The observations of a sudden increase and then decrease of acetic acid content in leachate from the control unit was less dramatic with the recycle unit. Instead, the recycle unit performed more steadily than the control unit in terms of the volatile acid and pH changes, CH_4:CO_2 ratios, etc. This performance was less affected by single-pass washout and allowed the results from the recycle unit to be more predictable.

The iso-butyric acid of the control unit increased from zero to 1,000 mg/ℓ, and then decreased again to zero, whereas for the recycle unit, it increased from zero to 950 mg/ℓ, and then decreased to zero (See Figures 15 and 16). The corresponding concentration of propionic, butyric, and valeric acids vary in the ranges between the acetic and the isobutyric acids. As in other anaerobic fermentation processes, acetic acid is a major product produced by microorganisms in landfills. However, condensation reactions are possible which form other acids of higher molecular weight, such as butyric and valeric acids. The odd numbered fatty acids, such as propionic and valeric acids could also be formed by such reactions. (11, 48, 49). The ultimate variation of the volatile fatty acids is dependent upon the pH, ORP and other factors. In the early stages of decomposition, fatty acids are abundant, the pH will likely be in the acidic range (approximately 5.0), and ORP values may be less negative. As the landfill matures the conditions may reverse.

Membrane ultrafiltration (UF) and gel permeation chromatrography (GPC) were performed to separate and determine the classes of organics present in the leachate. Prior to the lab-scale landfill study, a preliminary evaluation of these processes had been conducted, so that the performance and quality assurance can be established. (see Appendix IV)

A 200-ml sample of both the control and recycle leachate was filtered and TOC values were then determined. An Amicon UM05 (nominal molecular-weight cut off of 500) UF membrane was used to concentrate the high-molecular-weight compounds. The TOC of the retentate and permeate were again determined (see Table 13). For a two-fold concentrated solution (i.e., samples collected on 5/25/81 and 6/16/81), the same TOC concentration was found in both the retentate and permeate solutions (see Table 13). This indicated that the organics present in the leachate consisted mainly in the low-molecular-weight range (e.g., less than 500). This result confirmed the observation that the volatile fatty acids were important constituents of the leachate samples from both units, especially when high TOC concentrations prevailed. (see Figure 15 and 16)

Table 13 indicates that the recovery of TOC by the UF membrane is always lower with the leachate from the recycle unit. Apparently there were more low-molecular-weight organic compounds, such as the volatile fatty acids, present in the leachate samples from the recycle unit. This implied that the recycle operation provided better opportunity for degradation of the more readily biodegradable organics in the leachate from that unit than the leachate from the control unit.

The retentate was further separated using a GPC column. The results of the GPC runs are shown in Table 14. Approximately 84% of the organics (based on TOC value) was in the low-molecular-weight fraction.

TABLE 13. Concentration of TOC in the Retentate and Permeate of the UF Process.

Sample Date	Concentrating* Factor	Original, mg/l		Retentate, mg/l		Permeate, mg/l		Recovery, %**	
		C	RC	C	RC	C	RC	C	RC
5/25/81	2.0	7000	11200	7600	7200	6800	7200	54.3	32.1
6/16/81	2.0	9800	11700	8900	9800	8900	9630	45.4	39.7
7/13/81	4.0	10679	11093	6209	11589	6954	8444	14.5	26.1
8/17/81	4.1	2900	9000	6740	12100	1230	7500	58.1	33.6

* Concentrating factor = $\frac{\text{Original Volume, (ml)}}{\text{Final Volume, (ml)}}$, where the original volume was 200 ml.

** Recovery, (%) = $\frac{\text{Retentate TOC, (mg)}}{\text{Original TOC, (mg)}} \times 100\%$

TABLE 14. Concentration of TOC of Each Molecular Weight Fraction of the Leachate Retentate.

Sample Date	Total, mg/l		High[1], mg/l		Medium[2], mg/l		Low[3], mg/l	
	C	RC	C	RC	C	RC	C	RC
5/25/81	6390	7470	570	230	140	360	5680	6880
6/16/81	8530	6700	840	880	560	190	7130	5630
7/13/81	5663	7202	1225	332	464	579	3974	6291
8/17/81	2490	7295	438	1175	495	410	1557	5710

Note: 1. "High" means the fraction with M.W. > 50000

2. "Medium" means the fraction with M.W. 1000 < M.W. < 5000

3. "Low" means the fraction with M.W. < 1000

pH is also an important parameter for an anaerobic landfill. The pH value is an important determinant in buffer capacity considerations. It also reflects the variation of the volatile fatty acids (acetic, propionic, butyric, and valeric acids) in the leachate. Therefore, a decrease in these volatile acids should be accompanied by an increase in pH. The low pH values (5.2 to 5.7) in the first 62 days for the control unit and in the first 110 days for the recycle unit, as shown in Figure 18, indicate that the low-molecular-weight volatile fatty acids (mainly acetic acid) were prevalent in the leachate during that period. Gas chromatographic analysis of leachate samples for the specific volatile fatty acids (see Figures 15-17) confirmed these results.

The pH of both the control and the recycle units increased from an initial of 5.35 to 7.26 and 5.25 to 7.16, respectively. The pH of the leachate samples collected from the control unit increased rapidly to a neutral value of 7 during a two-week period (Days 62 to 76 in Figure 18), whereas that from the recycle unit increased gradually to a neutral value of 7 during a seven-week period (Days 92 to 141 in Figure 18). This implied that the predominant microorganisms in the simulated landfills shifts from acid formers to methane formers. These changes in pH correspond to a buffer shift from that established by the volatile acids to that of the more favorable bicarbonate system that is also considered necessary for efficient CH_4 production. This shift is shown also by decreased in conductivity (see Figure 21) as well as in total alkalinity attributable to the volatile acids. (Figure 19)

The alkalinity of the leachate sample provides the buffer capacity of the system and as shown in Figure 19, alkalinity of the control unit increased from 4,250 mg/ℓ to 7,150 mg/ℓ, and then decreases gradually to 4,130 mg/ℓ, whereas that of the recycle unit rose from 4,500 mg/ℓ to

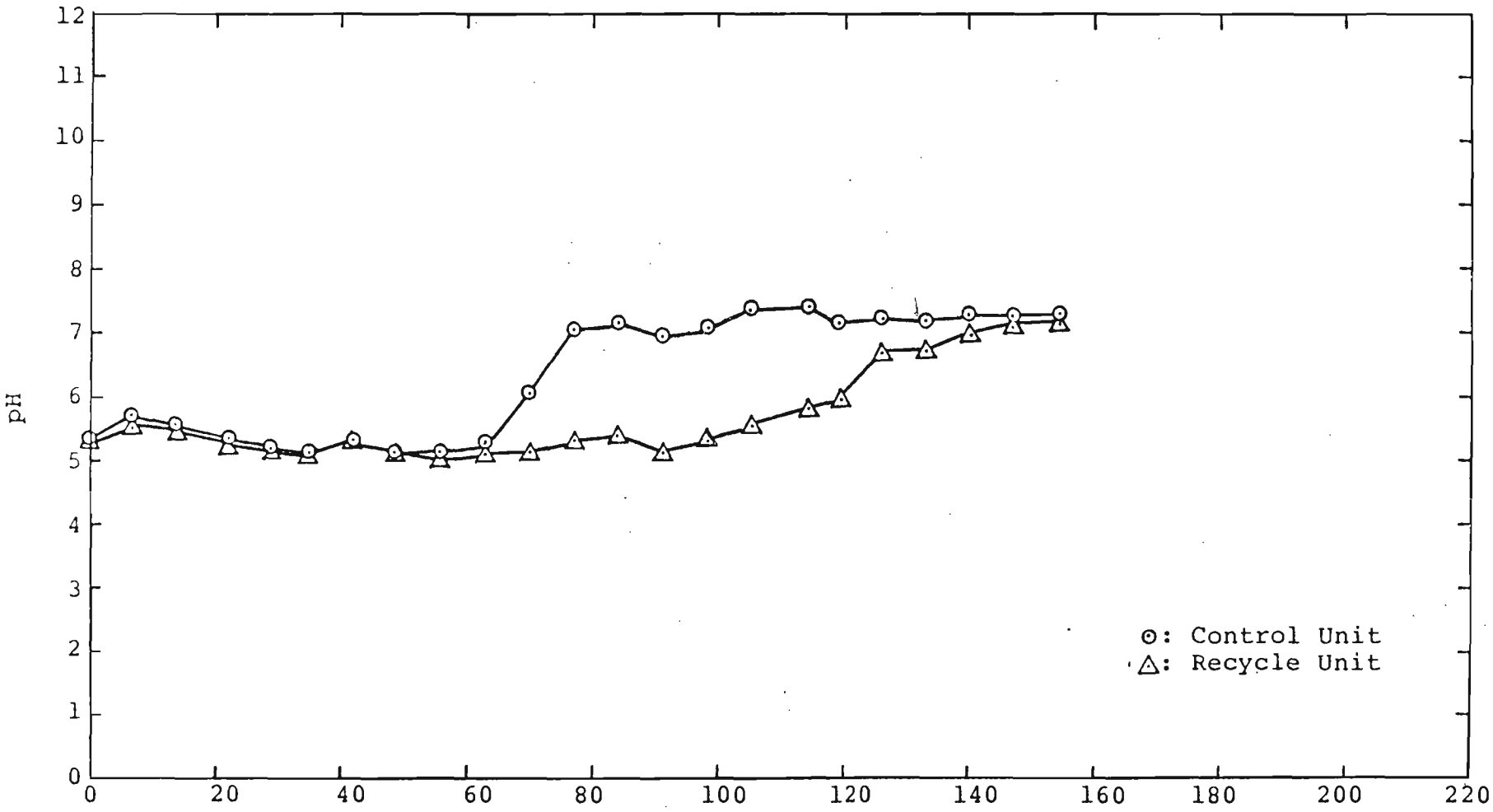

Figure 18. pH of Leachate

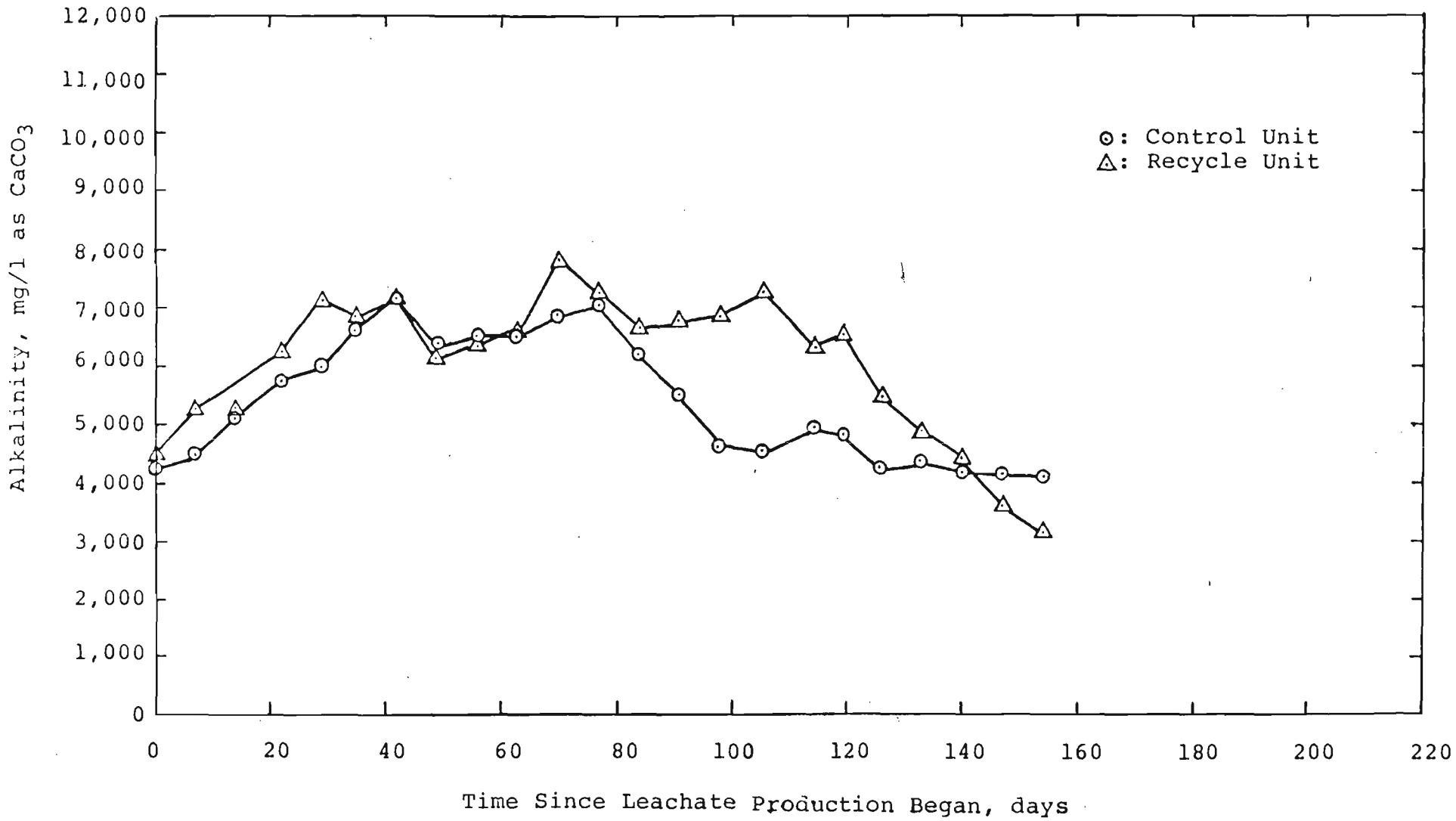

Figure 19. Alkalinity of Leachate

7,800 mg/ℓ, and then decreased to 3,180 mg/ℓ. The increasing and decreasing in alkalinity were due to the shifting of buffer system. The accumulation of volatile acids correspond to the increase of alkalinity, and later on as volatile acids were converted to CH_4 and CO_2 by the biological populations, caused a decrease in alkalinity and a shifting buffer capacity to the domain of bicarbonate system.

The ORP of leachate from the control unit reduced from 10 to -233 mV, whereas that of the recycle unit from 35 to -233 mV (see Figure 20). The eventual low ORP values indicated that both units were operating under highly reduced conditions. Sulfates and sulfites in leachate are transformed to sulfides under these conditions. A dark-green/black-colored and rotten-egg-smelling leachate was produced for both units and sulfides were also produced in the leachate from both units. The low ORP values also indicated that these landfill units were being operated under conditions conducive to anaerobiosis (11).

The conductivity of the leachate samples from the control unit increased from 7,600 μmhos/cm to 15,500 μmhos/cm and then decreased to 8,900 μmhos/cm, whereas that of the recycle unit increased from 8,000 μmhos/cm to 15,800 μmhos/cm, and then decreased to 7,000 μmhos/cm (see Figure 21). A ultimate lower conductivity values obtained in leachate samples from the recycle unit indicated that there were less dissociated ions in the recycle unit leachate than in the leachate from the control unit.

The increasing in conductivity was due to the formation of volatile acids, releasing of inorganic and organic materials by the biological activities in early stages of landfill. When landfill approaches stabilization, less organic materials was presented in leachate and the metal ions were precipitated and filtered out by sulfide under a reduced chemcial condition, (i.e., with ORP lower than -200 mV), whence, a decreasing in

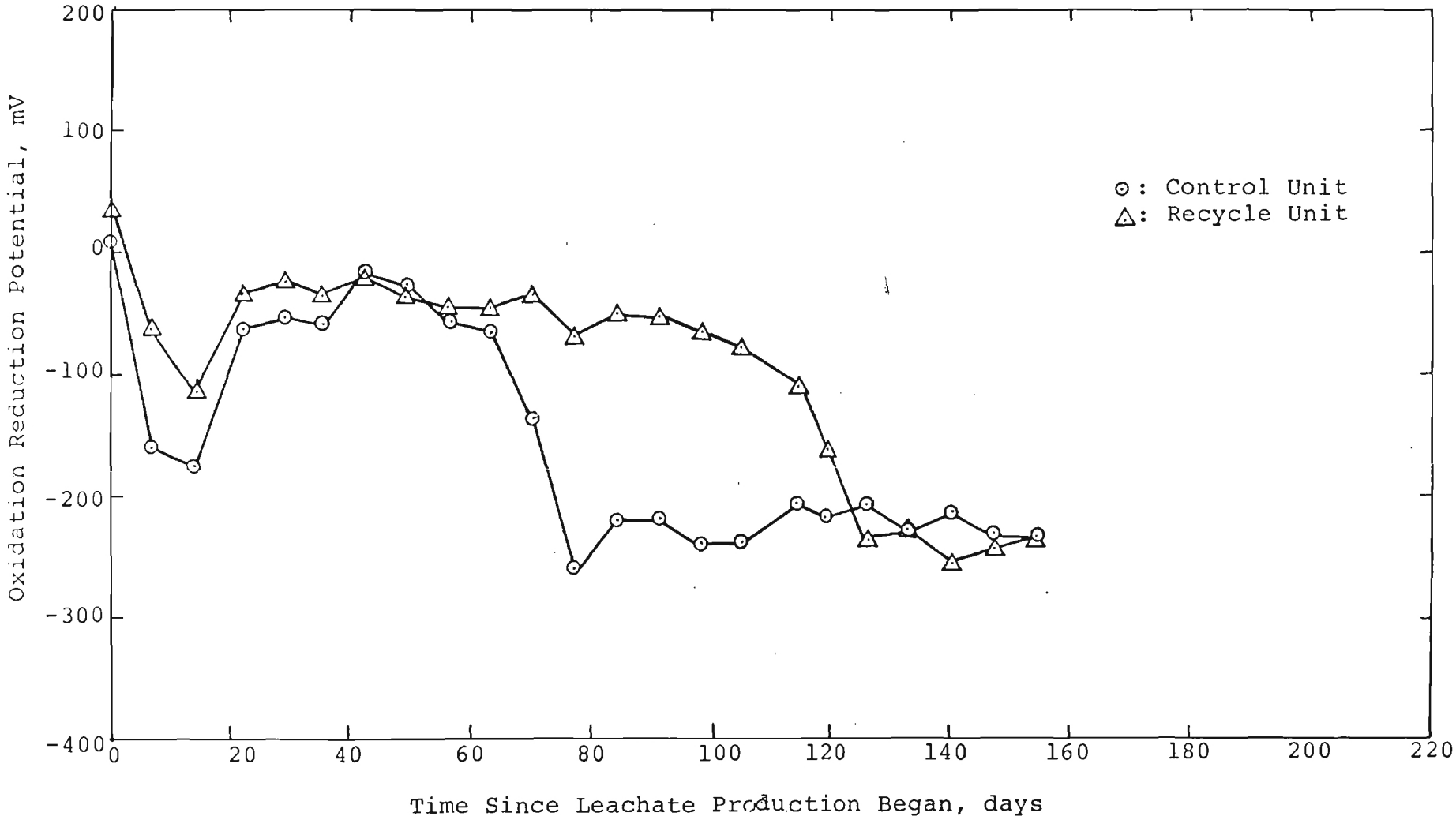

Figure 20. Oxidation Reduction Potential of Leachate, days

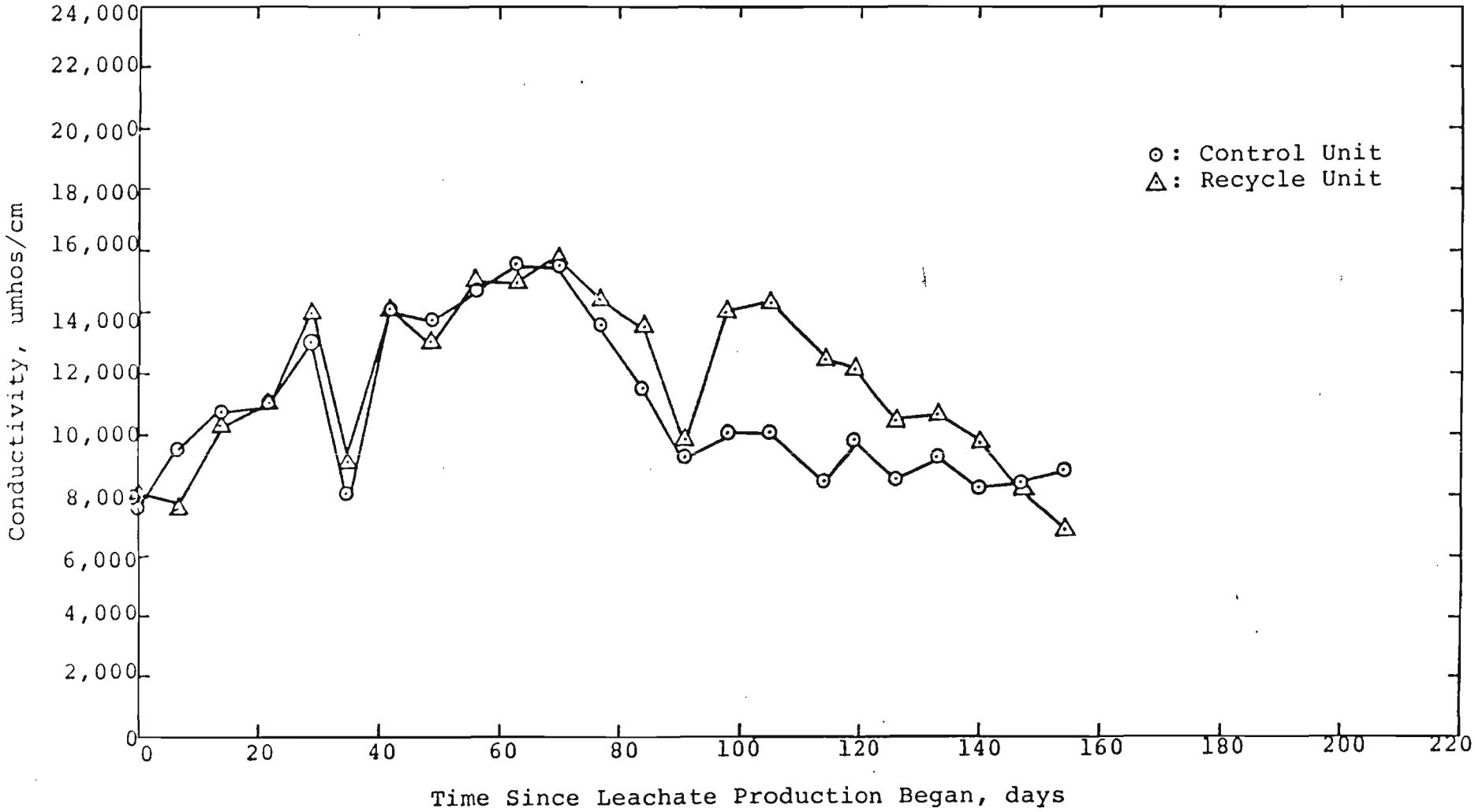

Figure 21. Conductivity of Leachate

conductivity was observed. The washout effect of the control unit also contribute to the decreasing of conductivity.

The variations of the radionuclides, i.e., Co-58, Sr-85, Cs-134 and H-3 are shown in Figure 22. No significant changes in activities were observed in the first two months for both units. However, in the third month, the activity of Co-58 in the leachate of the control unit decreased abruptly from 1.0 to 0.05 μCi/ℓ. During the same period, the corresponding ORP of the control unit decreased below -200 mV (Figure 20). At the same time, sulfide concentrations increased from zero to a peak of 3.6 mg/ℓ (see Figure 23). The same low values of Co-58 activity were observed in the leachate from the recycle unit (Figure 22). However, it took an additional six weeks to attain a peak sulfide concentration of 6.7 mg/ℓ (Figure 23). Apparently decreased in Co-58 activity were related to the presence of sulfides which would promote precipitation reactions.

The decrease of Co-58 activity in the leachate of the recycle unit was smoother than in the control unit. This appears to agree with the gradual changes of other parameters, such as pH, ORP, etc. and the greater leachate homogeneity encouraged by the use of leachate recycle as opposed to single-pass operation. The activities of other radionuclides, however, only decreased slightly and were apparently not greatly influenced by the presence of sulfides. This may have been due to the high solubility of the isotopes, i.e., Sr-85, Cs-134, and H-3, in leachate and a lesser opportunity for interaction.

The activity of Sr-85 in the leachate of both units decreased from 6.0 to 1.0 μCi/ℓ, which might be due to the formation as a insoluble carbonates. The activity of Cs-134 also showed one order of magnitude decreasing (from 1.2 to 0.12 μCi/ℓ), which might be due to the adsorption/ion-exchange mechanism of the trace amount of soil presented in solid waste.

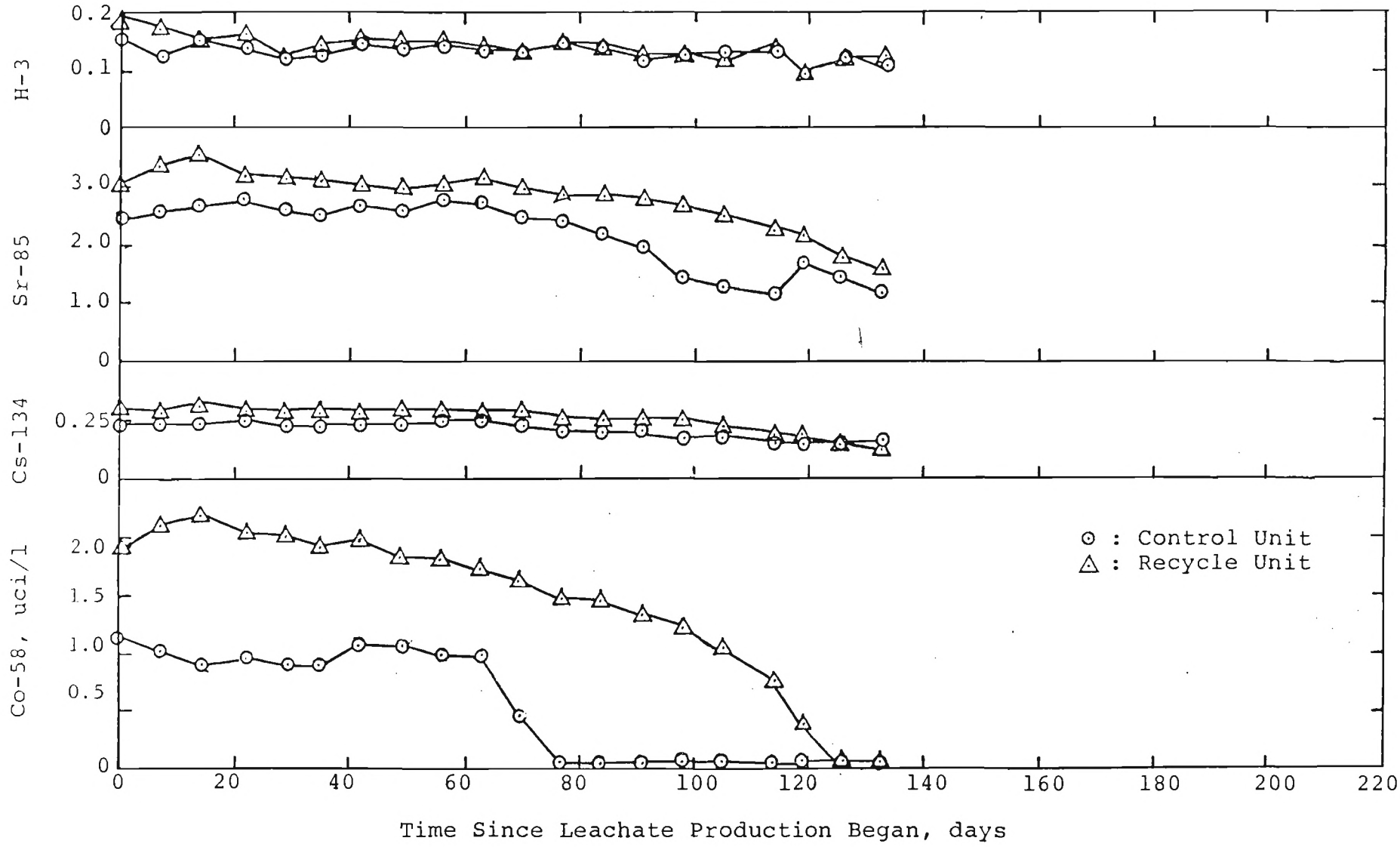

Figure 22. The Activity of Radionuclides in Leachate

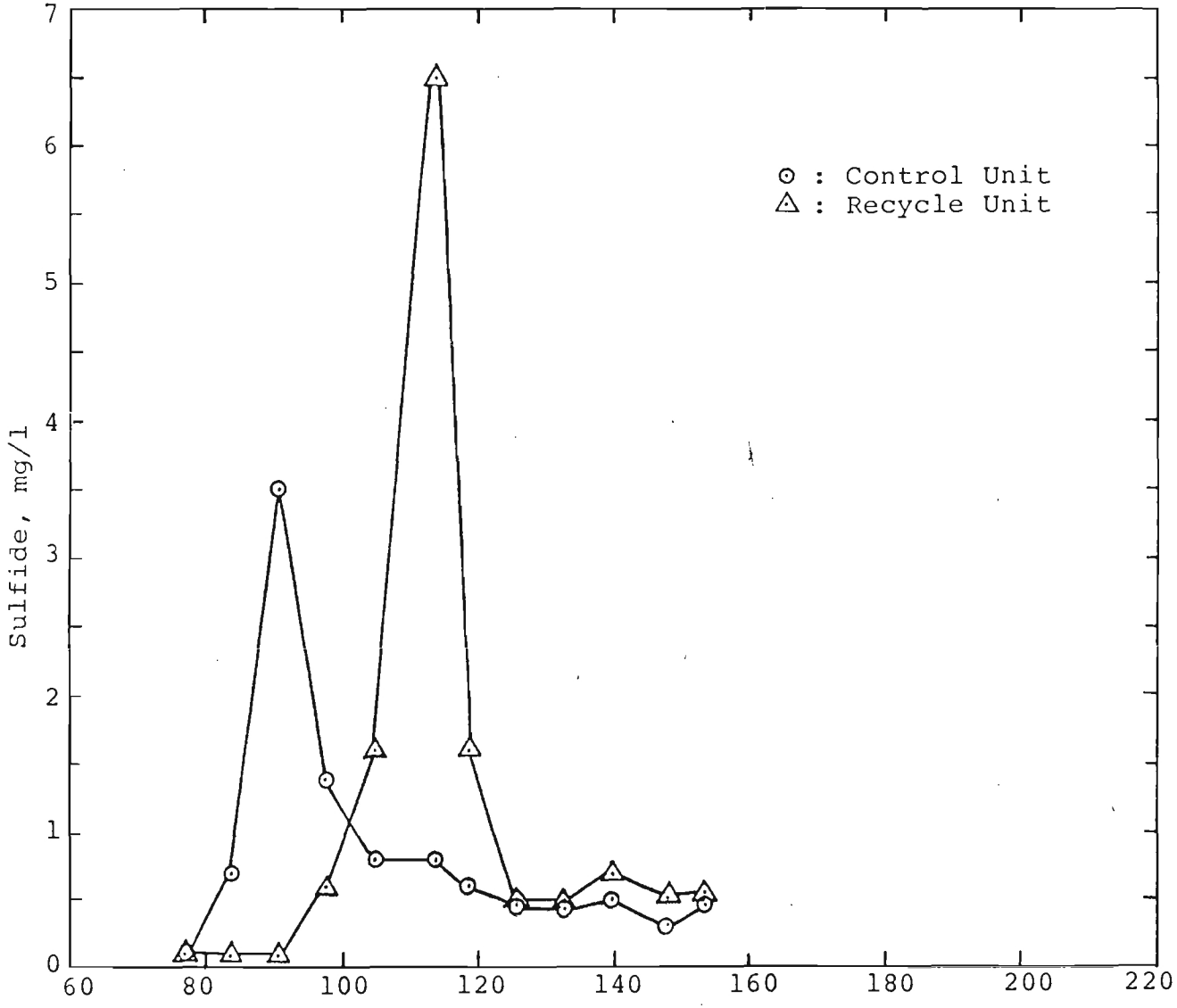

Figure 23. Concentration of Sulfide in Leachate

The concentrations of non-radioactive Co and Cs are listed in Table 15. The same decreases in Co concentrations were observed. Again, the availability of sulfides in the leachates from both units (Figure 23), with highest concentrations being 3.5 mg/ℓ on Day 92 and 6.5 mg/ℓ on Day 112 in the control and recycle units, respectively, probably caused this decrease and removal as metal sulfides.

Six other metals were examined in this study, including iron, potassium, sodium, manganese, magnesium, and calcium. The variations of these metals in the leachate samples are shown in Figure 24, 25, and 26. The fluctuations of many of these metal concentrations are also found to closely correspond to those of sulfide and other parameters, e.g., pH, ORP, conductivity, etc. (Figures 18-23).

The removal mechanisms of these metals are mainly physical-chemical processes (4, 29, 39, 50), the biological processes also contribute to a lesser extent. Lower concentration of these metals in leachate of the control unit should be due to the wash-out effect.

The results of total suspended solids (TSS), volatile suspended solids (VSS), and the ratios of VSS/TSS in the leachate samples are depicted in Figures 27, 28 and 29, respectively. It can be seen in Figure 27 and 28 that both TSS and VSS of these leachate samples collected from the recycle and the control units show a slight initial decrease, and then increased dramatically to a maximum on the 77th day for the control unit and on the 123rd day for the recycle unit. However, peak values of TSS and VSS from the recycle unit lasted for approximately 15 days whereas those from the control unit lasted only a day or so. In both cases these values (TSS and VSS) returned to the original concentrations observed prior to peaking.

The sudden surge of TSS and VSS in the leachates can be interpreted by comparison with the respective ORP values (see Figure 20) which show values

TABLE 15. Concentration of Non-Radioactive Co, Cs and Sr of Leachate

	Co, mg/l		Cs, mg/l		Sr, mg/l*	
Sample Date	C	RC	C	RC	C	RC
5/18/81	0.095	0.162	0.030	0.040	-	-
6/1/81	0.082	0.162	0.025	0.025	-	-
6/16/81	0.142	0.090	0.030	0.035	-	-
6/29/81	0.115	0.090	0.030	0.015	-	-
7/13/81	0.105	0.125	0.040	0.025	-	-
7/27/81	0.077	0.147	0.025	0.030	-	-
8/10/81	0.050	0.137	0.030	0.040	-	-

*Due to the inconsistency of the instrumental output, no data was available to date.

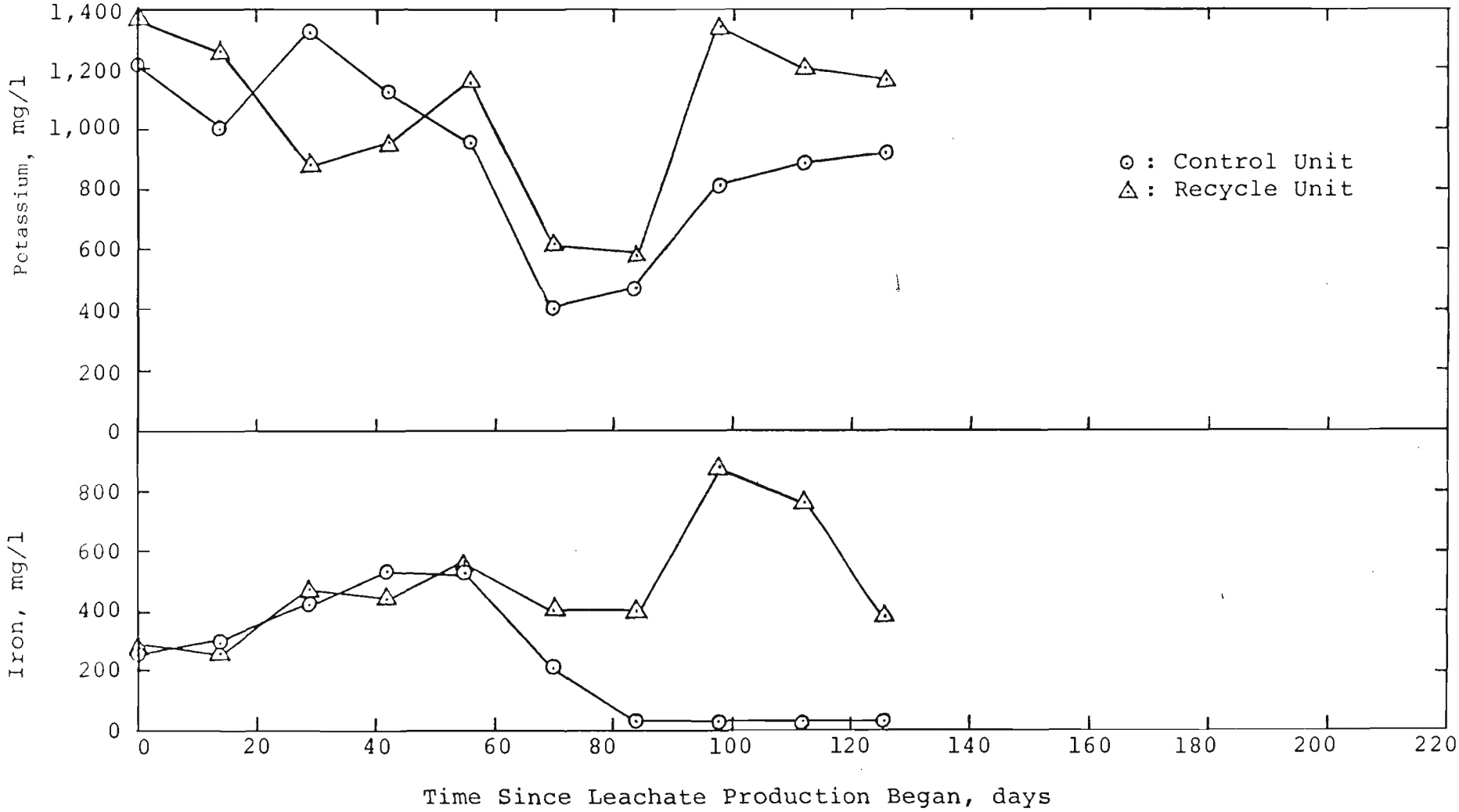

Figure 24. Concentrations of Iron and Potassium in Leachate

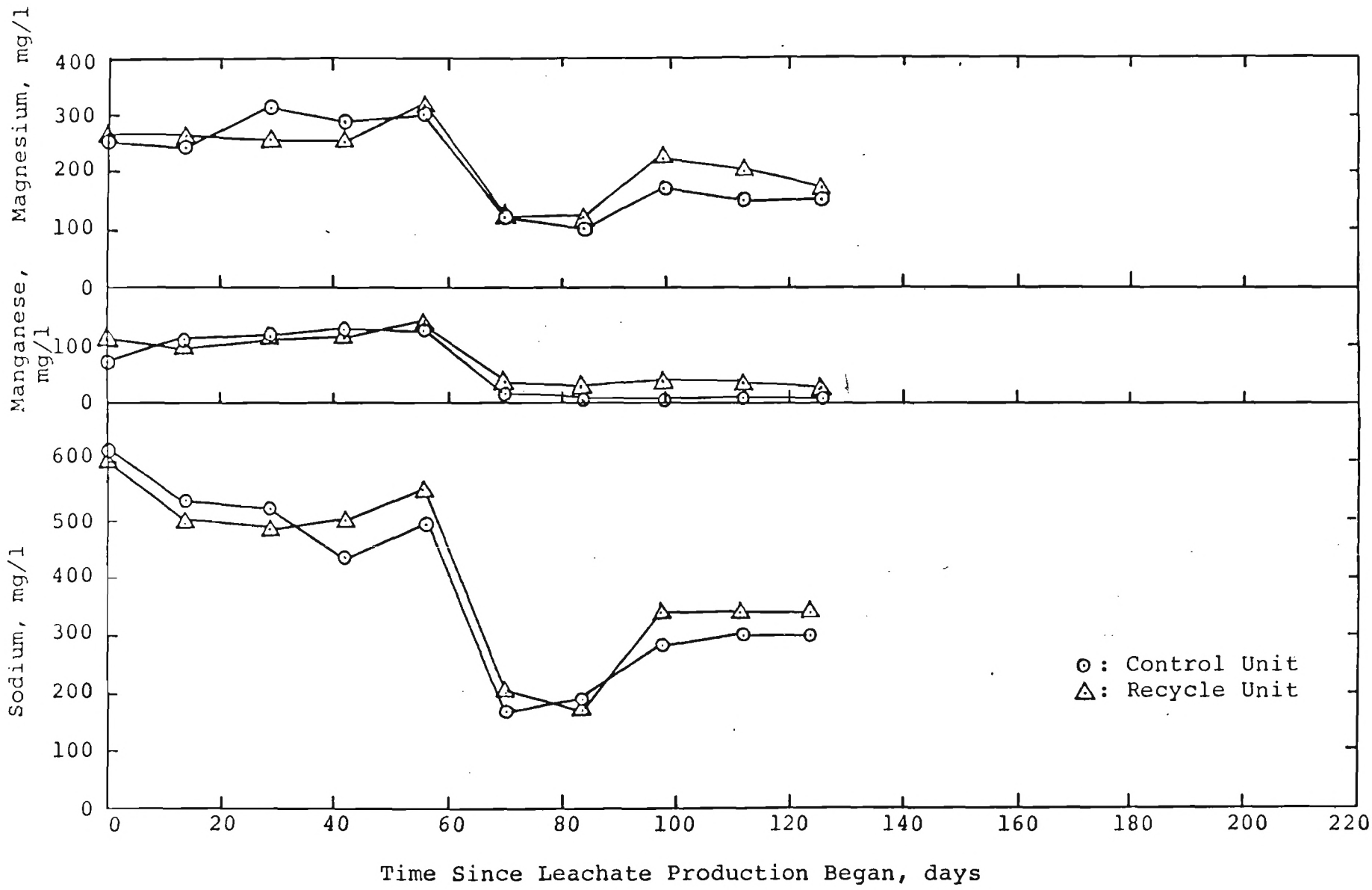

Figure 25. Concentrations of Sodium, Manganese and Magnesium in Leachate, mg/l

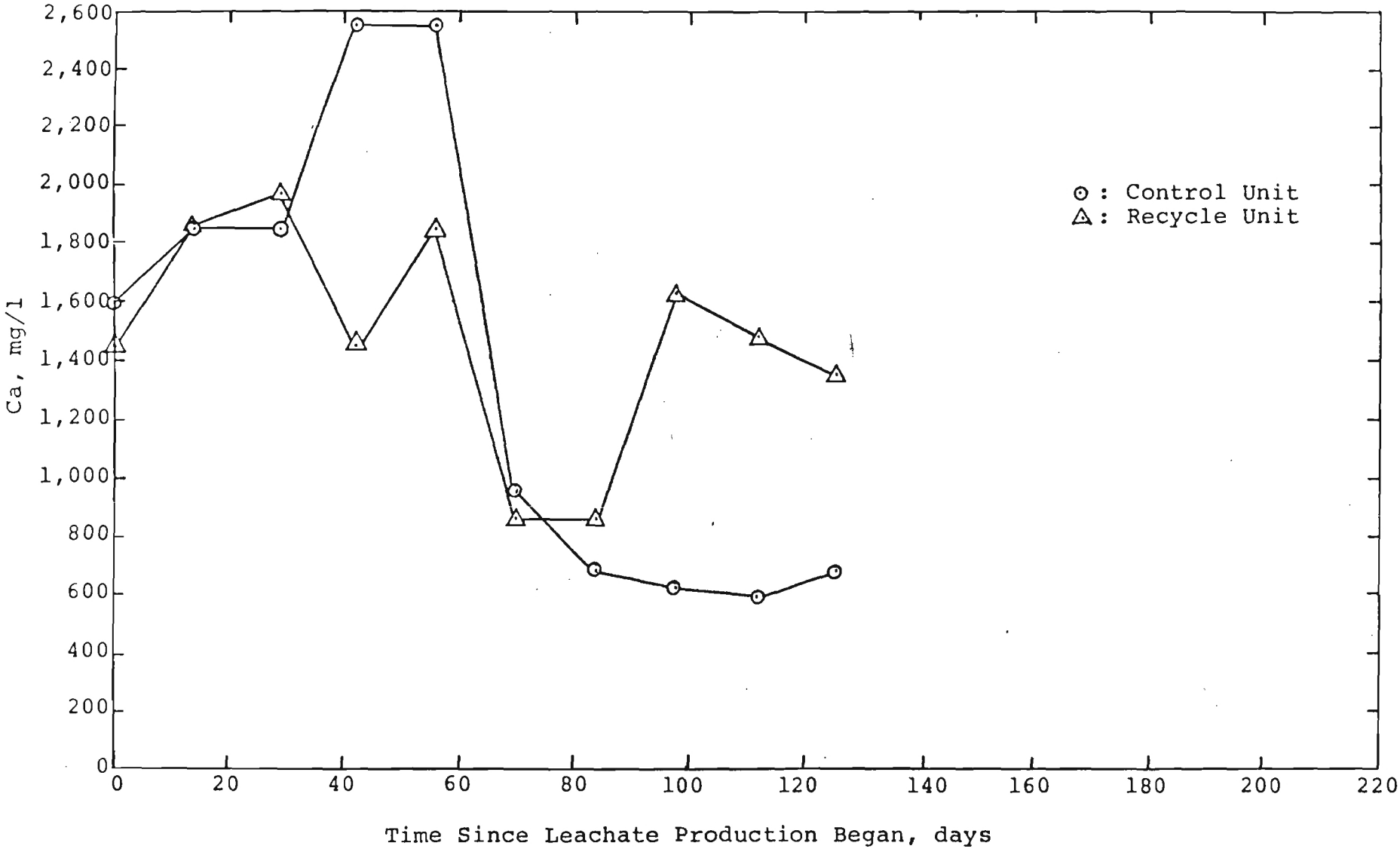

Figure 26. The Concentration of Calcium in Leachate

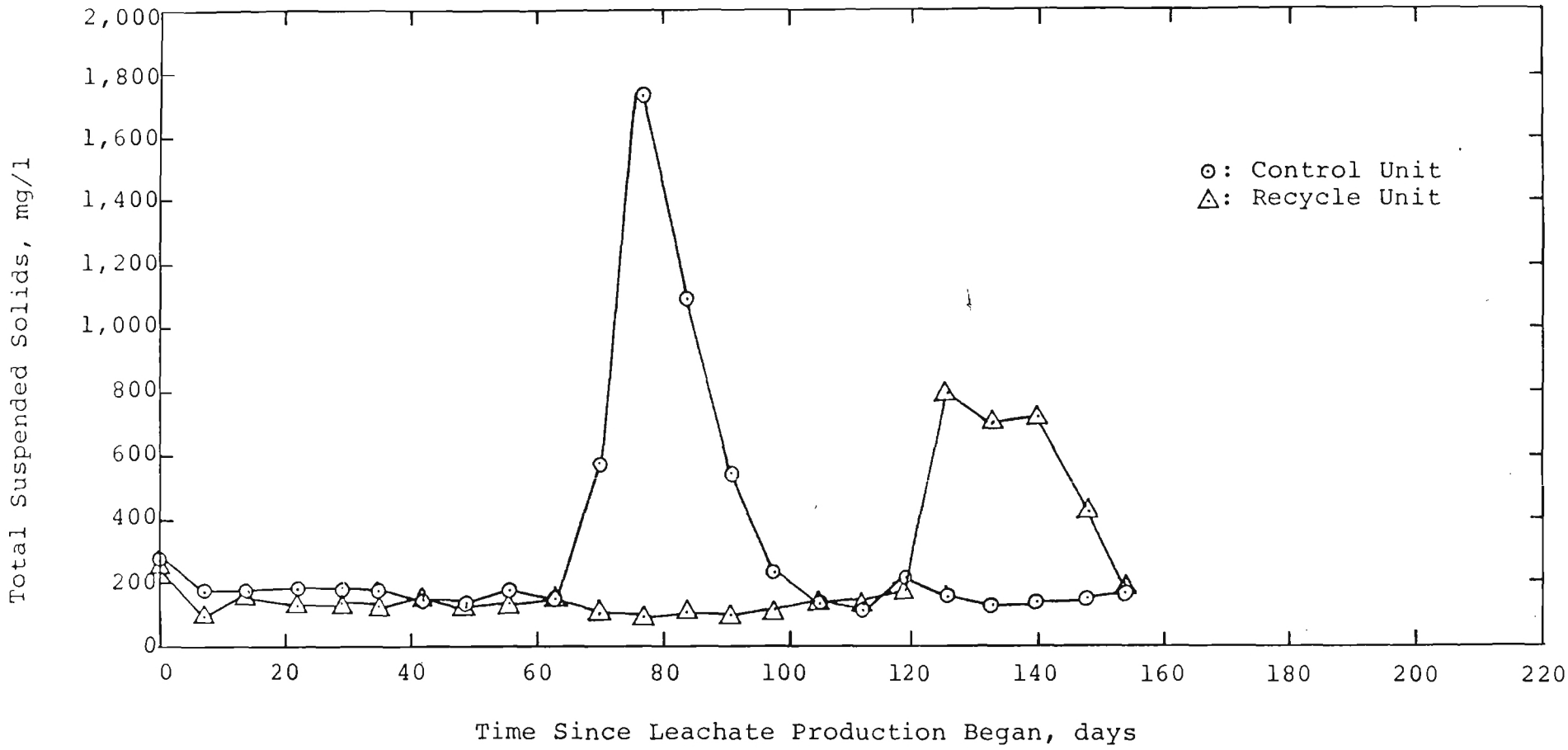

Figure 27. Concentration of Total Suspended Solids in Leachate, mg/l

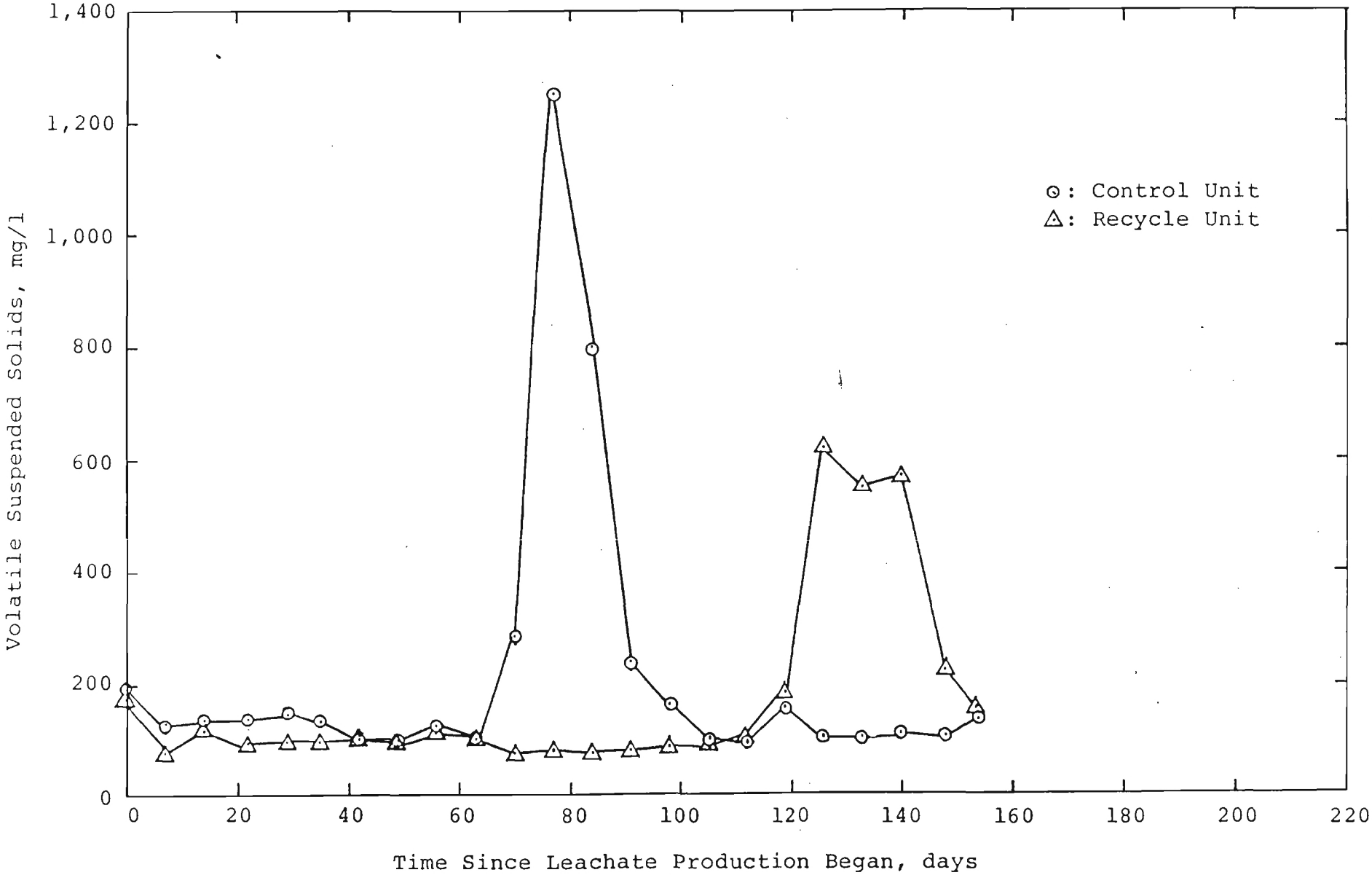

Figure 28. Concentration of Volatile Suspended Solids in Leachate, mg/l

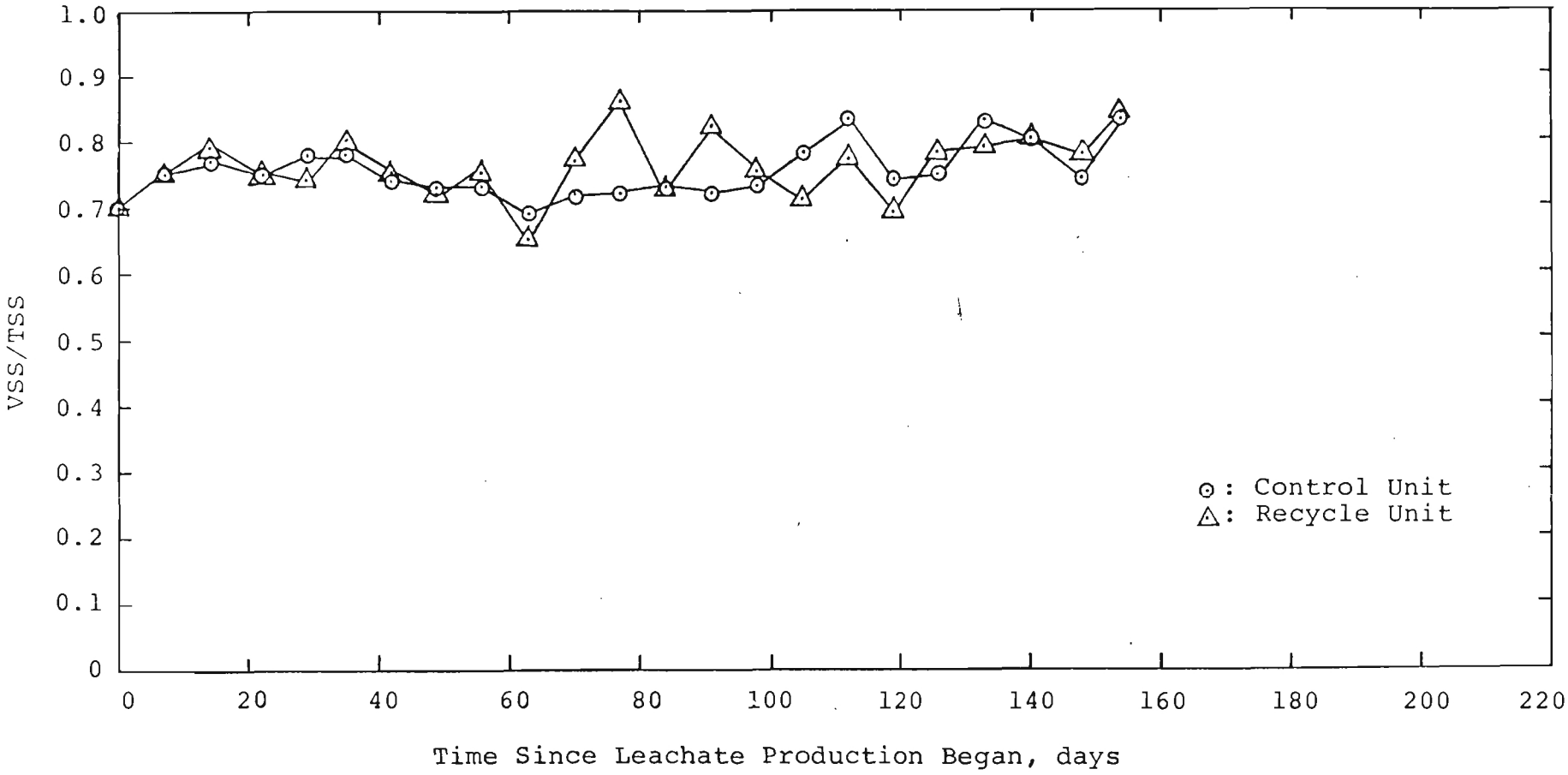

Figure 29. Ratio of VSS/TSS in Leachate

lower than -200 mV on the 78th day for the control and on the 123rd day for the recycle unit. Under such a highly reduced condition, sulfide formation was likely initiated. The subsequent precipitation of the sulfide compounds would tend to cause a sudden surge in the TSS and VSS concentrations in the leachate samples. These coincide with a significant drop in the metal concentrations of the leachate samples as discussed earlier.

Figures 27 and 28 also indicate that the steady-state values of TSS and VSS in leachate collected from the recycle unit are consistently lower than those from the control. It appears that both TSS and VSS in the leachate were being constantly filtered from the leachate each time the leachate was recycled through the refuse. The fact that the maximum concentrations of TSS and VSS in the leachate from the recycle unit were also lower than those from the control unit also supports this premise.

Figures 27 and 28 also indicate initial TSS concentrations of 274 and 251 mg/ℓ, respectively, for the control and the recycle units, whereas maximum TSS concentrations were 1730 and 800 mg/ℓ, respectively. Moreover, the TSS and VSS concentrations from the control unit fluctuated somewhat more than that of the recycle unit.

3.5 Gas Production

The gas production rate and its composition reflect the relative biological activity within the landfills. They also provide information regarding the intrinsic roles of acid and methane formers during the course of anaerobic stabilization.

The cumulative amount of gas produced and its composition from the control and the recycle units are shown in Figures 30, 31 and 32, respectively. The amount of gas produced from the recycle unit increases slowly in the first three months, and then rapidly afterwards. The maximum

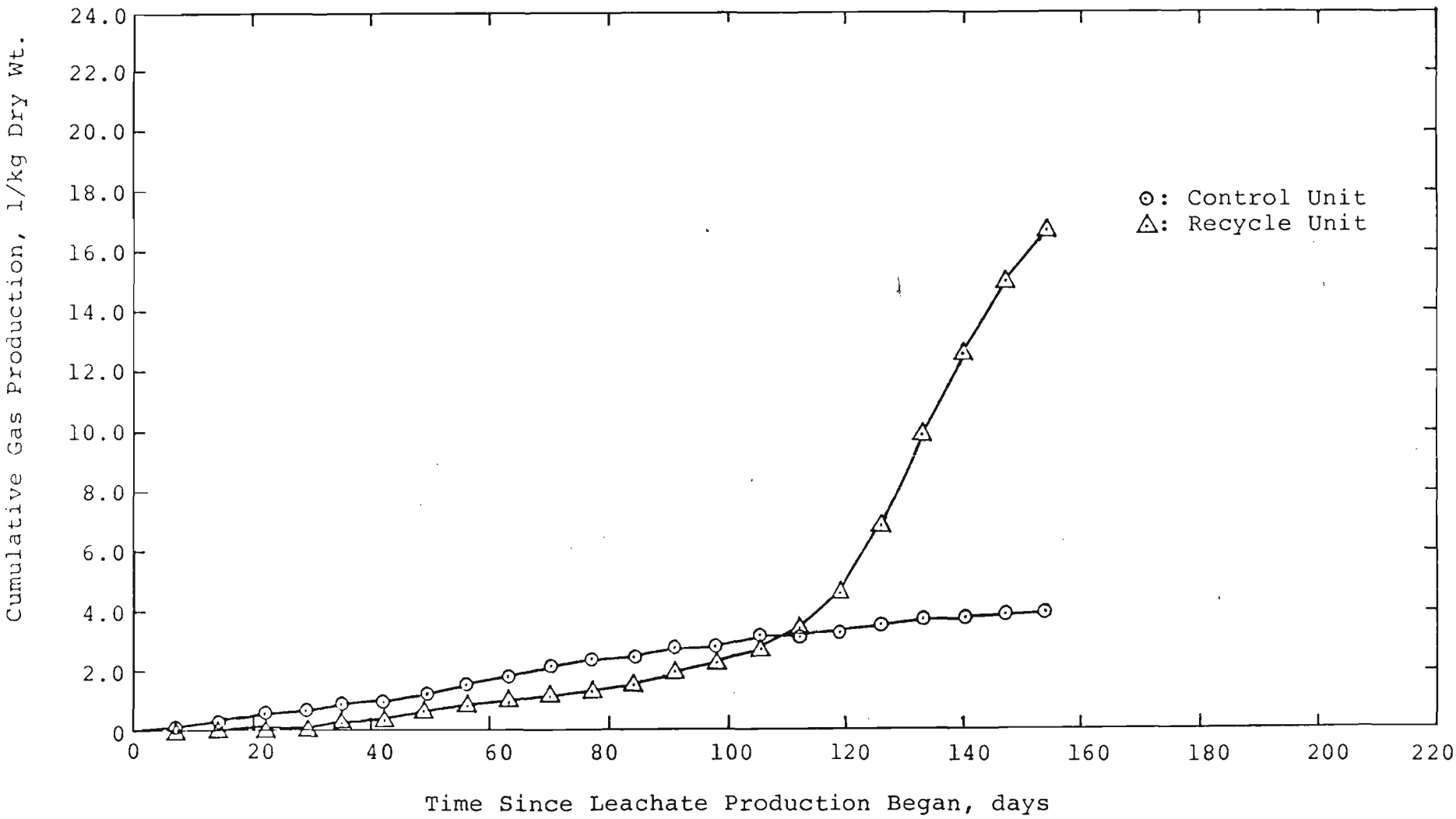

Figure 30. Cumulative Gas Production

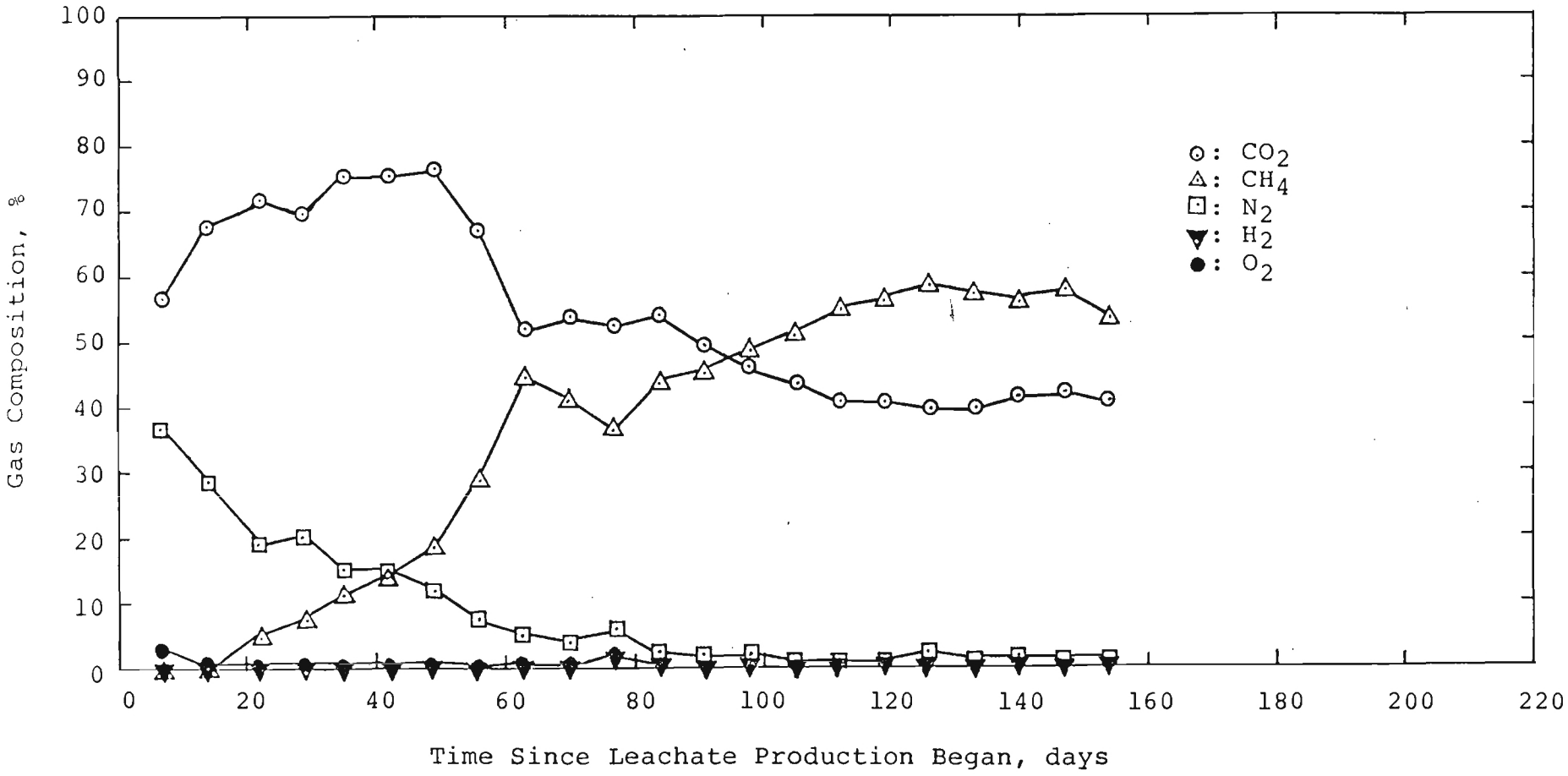

Figure 31. Composition of Gas from the Control Unit

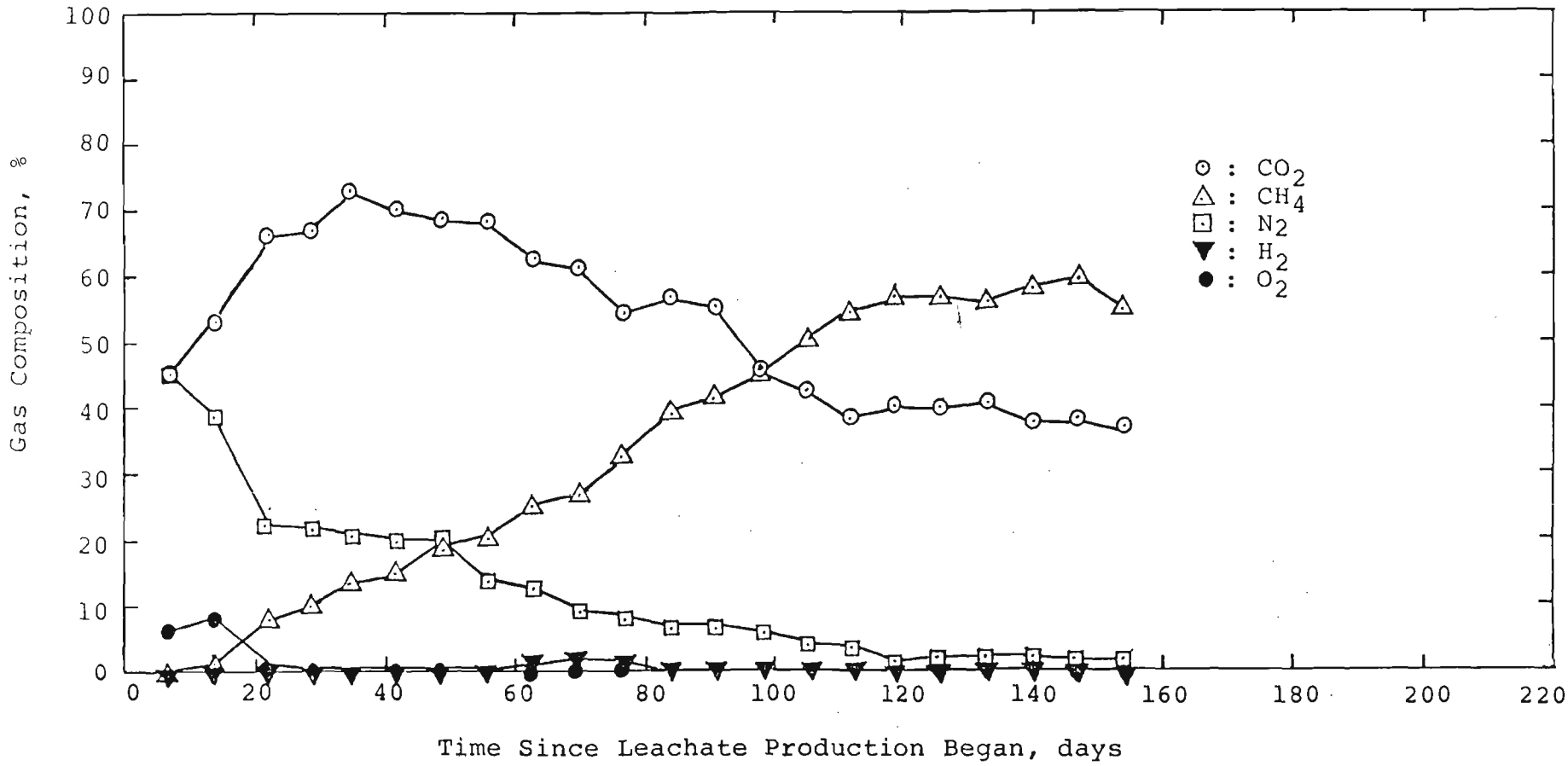

Figure 32. Composition of Gas from the Recycle Unit

gas production rate is less than 2 ℓ/day (including gas produced from the leachate reservoir*). The cumulative amount of gas produced from control unit was 3.8 ℓ/kg dry wt. on Day 158 as compared to 16.7 ℓ/kg dry wt. of solid waste obtained with the recycle unit (Figure 30). Based on the amount of gas produced from the two units, it can be concluded that the recycle unit had more rapidly and completely stabilized organic constituents than the control unit. These results confirm the earlier observations of Pohland and coworkers (49).

As can be seen from Figures 31 and 32, the onset of methane generation from the control unit occurred one week later than from the recycle unit. The generation of hydrogen from the control was two weeks later than the recycle unit. Since the formation of CH_4 and H_2 can be taken as indicators of the degree and stage of stabilization at a landfill site (16, 59), it was expected that the recycle unit would provide leachate stabilization sooner than the control unit.

The carbon dioxide composition of the gas from both units increased to a maximum and then decreased (see Figures 31 and 32). The maximum CO_2 values for the control and the recycle units were 76.5% and 73.2%, respectively, whereas the ultimate CO_2 values were 40.2% and 36.6%, respectively. The corresponding methane compositions increased from 0% to 53.4% for the control, and from 0% to 55.5% for the recycle unit. A

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*In the early stages of operation, gas produced from the leachate reservoir of the control unit was connected to the same gas collecting buretts for the landfill unit. This arrangement led to a faulty indication of net gas production and composition from the control unit. System alterations were made on the 70th day of the research to separate the respective gas collecting systems from the reservoir and landfill unit of the control unit. It should be noted that data for the control unit shown in the first 70 days indicated the combined value of gas collected from both the reservoir and landfill unit.

normalized gas composition, based on CO_2 and CH_4, is shown in Figure 33. As could be expected, the variations in CO_2 and CH_4 from the more stable recycle unit were less than those of the control unit. The ultimate gas composition for the control unit was 43% CO_2 and 57% CH_4; for the recycle unit, 40% CO_2 and 60% CH_4. The amount of gas produced was normalized on a daily basis to the standard conditions, i.e., 0°C and 760 mm Hg. The vapor pressure of water was substracted from the total pressure prior to the normalization of the amount of gas produced.

The oxygen content in the gas from both units was nearly depleted within 30 days after initiation of operation. The nitrogen content of the control unit decreased from 38% to 1.5%, whereas that of the recycle unit decreased from 46% to 20% (see Figures 31 and 32).

Tritium was found in the gas produced at an activity level of 0.057 μCi/ℓ. The presence of tritium in the form of methane and water vapor is resulted from the biological conversion processes. Due to the detection of H-3 in the gas phase, the gas produced from the landfills must be treated before release to the atmosphere. Gas treatment can be provided by the procedure illustrated in Section 2.2.2. The final product of the condensed water is then stored and treated as a low-level radioactive liquid waste.

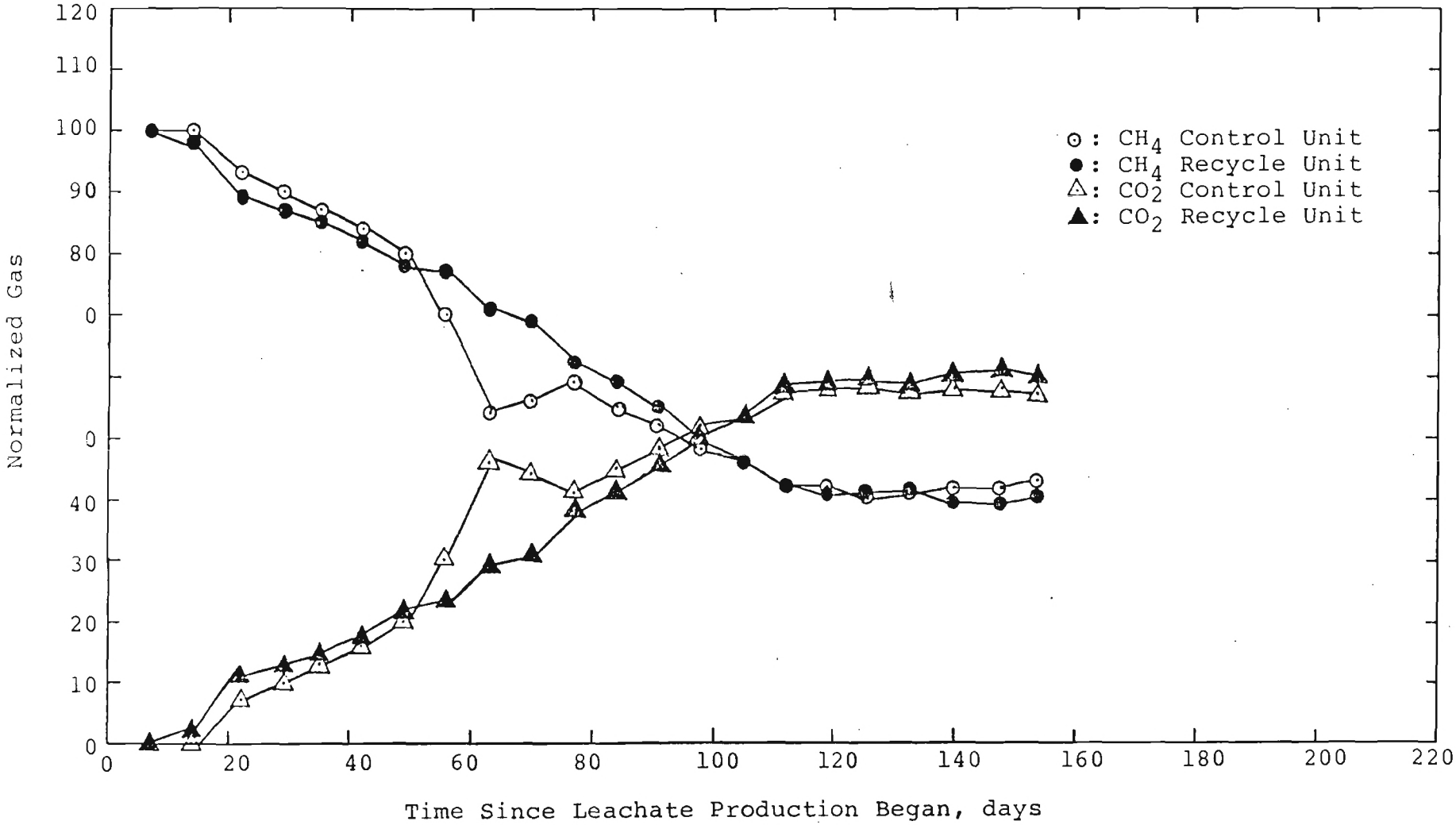

Figure 33. Normalized Composition of Gas from the Controlled and Recycle Units

IV. CONCLUSIONS

Based on the results of this study, the following conclusions can be drawn:

1. The more stable and predictable performance observed with leachate recirculation reinforced the value of this operational technique in landfill practice. Under the prevailing experimental condition, the time required for the simulated landfill with recycle to reach stabilization was approximately six months; that for the landfill without leachate recirculation has yet to be established and should be considerably longer.
2. Within the study, Co-58 was effectively contained in the landfill with leachate recycle, presumably in the form of metal sulfides. A two order of magnitude of Co-58 activity reduction was achieved in leachate (e.g., from 6.6 to 0.05 μCi/ℓ), whereas approximately one order of magnitude of activity reduction was attained with both Cs-134 (from 1.0 to 0.1 μCi/ℓ) and Sr-85 (from 6.0 to 0.8 μCi/ℓ). The degree of containment of these three radioactive isotopes in leachate was found to be Co-58 > Cs-134 > Sr-85. The mechanism of containing Sr-85 in landfill might be the formation of insoluble strontium carbonates and/or organic complexation effect, while that for Cs-134, sorption and/or ion exchange should play the major role.
3. Approximately 30% of tritium was found in the gases produced from the landfill units, and the remainder of 70% in leachate (e.g., 0.057 μCi/ℓ in gases and 0.12 μCi/ℓ)in leachate.

The presence of H-3 in gases should be a result of conversion of $^{3}H_{2}O$ into $C^{3}H_{4}$ by the biological activities. Therefore, the gases produced from landfills would require capture and treatment to eliminate radiation hazards from this source.

4. Based on this lab-scale study, co-disposal appears to be a promising and viable means of containing low-level radioactive wastes, particularly for those originating from small LLRW generation centers such as hospital, research institute, etc. In addition to the obvious advantages (such as more economical and convenient, less transportation risk, easier handling prior shipment, etc.) using the co-disposal method, leachate recirculation technique also permits the removal of radionuclides in a far better controlled manner.
5. The information obtained in this study would be useful in leading toward a more rational design of the pilot-scale landfill co-disposal study.

V. RECOMMENDATIONS

Based on the results of this preliminary study, the following recommendations are given:

1. A follow-up pilot-scale study of leachate generation and treatment during co-disposal of pertinent radioisotopes and municipal refuse is recommended using the following alternatives:

   (1) Leachate Recirculation. The leachate can be contained, collected and recirculated into the landfills without any further treatment. After the readily available organics are stabilized, the landfill can be drained and sealed. Accumulated residual leachate can then be subjected to ultimate disposal with or without further treatment. Addition of sulfates/sulfides or other complexing agents should be investigated during these studies to determine in situ treatment capabilities.

   (2) Off-site Treatment. The leachate generated from landfills can be collected and treated by: (a) physical-chemical processes, e.g., chemical precipitation, activated carbon and ion exchange adsorption, chemical oxidation, and reverse osmosis; and (b) biological process.

2. The gases generated from the landfill should be treated prior to its release into the atmosphere. One of the methods is to install a gas combustion equipment along with a scrubber for the removal of radioactive combustion products.

3. The design criteria for sanitary landfills receiving LLRW can be established based on the results gathered from a follow-up pilot-scale

study. This will include the dimensions of sanitary landfill, leachate recirculation rate, sulfate addition rate, liner materials, treatments of leachates and gases, sludge disposal, operation instructions and health and safety considerations.

4. Although pronouncing results were achieved in this lab-scale study, information on the use of other types of solid waste and LLRW, together with temperature effect, etc., is, however, limited. Therefore, a more comprehensive approach with the use of a larger-scale (i.e., pilot-scale) study should be conducted in the future.
5. The strategy for determining the specific treatment processes for any given radioactive isotopes should be established, regardless whether the isotope is applicable for co-disposal or not.
6. The safe guard for landfill sites when certain isotopes were presented in LLRW should be explored.
7. For a long term burial site, considerations should be given on design, containment, operation, management and closure procedures so that the most cost-effective means of disposing the specific LLRW can be achieved.

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Appendix I. Shake Down of the Lab-scale Landfill Units

Hydraulic and the pneumatic tests were conducted to locate possible leaks of liquid and gas from the lysimeters. The procedures for each text are detailed as follows:

1-1 Hydraulic Test

Hydraulic tests were employed to examine liquid seals for the entire system. The testing procedures were:

1. Fill the landfill unit and leachate reservoir with tap water to 4/5 of the tank height while keeping V-2, V-4, and V-5 closed (refer to Figure 8 for notations). Inspect for leaks which may occur during the first hour after filling with water. If there's any signs of leakage, drain the tank and seal the leaking points by tightening pipe joints or applying sealant. After that, refill the tank with tap water to the same water level, allow to stand for at least six hours, and observe any signs of minor leaks. Repeat the fill-drain-and-seal procedures for both major and minor leaks until no signs of leak are observed.

2. Fill the landfill unit with papers saturated with water and add tap water to 4/5 of the tank height (with V-2 closed). Proceed the hydraulic test as described in the previous paragraph.

3. Fill the landfill unit with papers saturated with water and add tap water to 4/5 of the tank height (with V-2 closed). Then, close V-3 and keep the rest of the valves opened. Examine any signs of leaks along the pipe line, including the pH/ORP measuring loop. Test the solenoid valve, along with sump pump, to see whether they function well or not.

1-2 Pneumatic Test

The pneumatic test was employed to examine for gas leakage from the entire system. The testing procedures are:

1. Fill the landfill unit and leachate reservoir with tap water to 4/5 of the tank height while keeping V-1, V-2, V-4, V-5, and V-6 closed.

2. Introduce 4 liters of N_2 gas into the landfill unit through the spare port, and two liters of N_2 into the leachate reservoir through the 0.9cm gas inlet. Record the temperatures of the room and tanks.

3. Let the system stand undisturbed overnight, then measure the gas volume collected in the burets. Record the temperature of the room and landfill unit.

4. Correct the gas volumes to standard conditions, and determine whether there is any change in the gas volumes.

5. If a significant loss of gas is observed through the system, introduce pressurized N_2 gas into the tank (with V-11, and V-12 closed) and detect areas of gas leaks by listening to the buzzing sound. Seal with a sealant or tighten the pipe joints if necessary to seal the leaks.

6. Repeat steps 3, 4 and 5 until no more loss of gas is observed

Appendix II. Filling and Spiking Method for the Landfill Units

The filling of the simulated solid wastes, and the spiking of the radionuclides for the lab-scale landfill units were conducted as follows:

1. The radionuclides (Co-58, Sr-85, Cs-134, H-3) were first dissolved in 0.1N HCl to a final volume of 200 mℓ, then stored in a polyethylene squeeze bottle.

2. The nonradioactive metals (Co, Sr, and Cs) were dissolved in distilled water to a final volume of 1 liter with a concentration of 750 μg/ℓ and stored in a glass bottle before use. After diluted in the landfill unit, a final concentration of 5 ppb was reached. The metal species were used as a tracer with respect to the radioactive form for the study of distribution between insoluble and dissolved forms.

3. The landfill units were filled with shredded solid wastes in six separate filling operations. The first five fillings had a depth of 10 cm; the last one, a depth of 5 cm. Between each solid waste layer, 1/5 of the reagents (as listed in Steps 1 and 2) were evenly spreaded onto the top of the solid wastes. The total depth of the solid wastes was then 55 cm.

4. Upon completion of filling and spiking the landfill units with solid wastes and metals, 52 ℓ of distilled water were pumped into the landfill units so that the field capacity of the solid wastes could be reached. As a result, the nonradioactive and radioactive metals (Co-58, Sr-85, Cs-134) and H-3 could be more evenly distributed with the landfill units.

Appendix III. Operating Procedures for the Lab-scale Landfill System

After the filling and spiking steps had been completed with the landfill unit, the entire system was operated as follows:

1. Turn off V-3, turn on V-2 and keep other valves opened.

2. After the first week of operation, 0.8 liter of leachate sample was collected from the landfill unit underdrain by shutting off V-4 and turning on V-3. Both the gross parameters and organic fractionation were performed after the radioactivity study had been conducted. Once these measurements had been completed, the contaminated samples (i.e., BOD_5, COD, and digested samples) were stored in a specific container for further treatment. The uncontaminated samples (i.e., the sample processed through Ge(Li), liquid scintillator, turbidity, heavy metal studies, and organic fractionation studies) were poured back into the leachate reservoir.

3. The gas sample was collected from the buret and analyzed for its composition and radioactivities. After analyses, the gas samples were stored in a tank for further treatment.

4. After the First sample, the sampling period for the rest was determined based upon the degree of variation of each individual physical/chemical and/or inorganic/organic component. The shortest sampling period was one week; the longest, two weeks.

Appendix IV. Characterization of Organic Matter in Leachate

The characteristics of the organic matter present in leachate can be taken as an indicator of the stages of decomposition and stabilization of a landfill (1, 2). When the low-molecular-weight organics, e.g., volatile fatty acids, predominate in leachate, the landfill is usually in its earlier stages of stabilization and most of the organics in the leachate are readily available for degradation by microorganisms. On the other hand, when the high-molecular-weight organics are predominant, the landfill is usually in its later stage of stabilization and the resulting leachate is less readily biodegradable.

The techniques for concentration and separation of organic matters in leachate have been well documented by Chian and DeWalle (3, 4). The concentration and fractionation processes employed in this study consisted of centrifugation, millipore filtration, membrane ultrafiltration, and gel permeation. A thorough evaluation of these processes prior to the lab-scale landfill study would not only minimize possible operating errors, but also verify the performance of each concentration and fractionation process as recommended by the instrument manufacturers. Evaluation of gross parameters (e.g., TOC, conductivity) according to Standard Methods (5) were used to monitor performance of these separation processes.

Leachate samples were collected from a simulated landfill cell used in a companion study on "Leachate Recycle as Landfill Management Option" (6). These samples were used to evaluate the proposed analytical scheme (see Figure 5 in Section 2.2.1.2). Two liters of raw leachate were collected in a glass jar (cleaned and baked overnight) and separated into two equal portions for analysis. Gross

parameter analyses were performed on the raw leachates, the results of which are given in Table 1. The low values of BOD_5 (16 mg/l) and ORP (-200.0 mV Ec) confirmed the fact that the leachate generated in this landfill sell was highly stabilized. Further evaluation of data in Table 1 showed that the COD/TOC and BOD_5/COD ratios were 1.27 and 0.04, respectively, indicating that the landfill had completed removal of much of the readily available organics.

The BOD_5, COD and TOC values of leachate after centrifugation, millipore filtration, membrane ultrafiltration, and gel permeation processes are given in Table 2. From inspection of Table 2, the centrifugation and MF processes did not significantly affect the BOD_5, COD, and TOC values of a stabilized leachate.

An Amicon Model TCF 10 Ultrafiltration Unit was used to separate high and low molecular weight fractions. Before the UF system could be used to evaluate leachate samples, the filter had to be tested for leaks (in terms of its molecular weight cutoff). Two compounds were chosen for this evaluation, i.e., Blue Dextron 2000 (high molecular weight compound) and sodium chloride (low molecular weight compound). A Beckman Model 26 spectrophotometer was used to monitor the Blue Dextran concentration (with wavelengths set at 619 nm). The NaCl was monitored with a Yellow Springs Instrument Model 31 Conductivity Bridge. The results of these studies are shown in Table 3. The results indicate that the high-molecular-weight compound is retained by the membrane (i.e., it is not present in the permeate and its value has doubled in the retentate), whereas the low molecular weight compound permeates the membrane (i.e., it is present in both the permeate and the retentate).

The millipore filtrate (400 ml) was concentrated two fold (volume

Table 1. Properties of Leachate from a Stabilized Landfill*

Parameter	Value
TC, mg/ℓ	544.0
TIC, mg/ℓ	192.0
TOC, mg/ℓ	352.0
COD, mg/ℓ	448.0
BOD_5, mg/ℓ	16.0
TSS, mg/ℓ	185.0
VSS, mg/ℓ	115.0
pH	6.5
ORP, mV Ec	-200.0
Conductivity, μmhos/cm	2,633.0

*The leachate used in this study was sampled from the same landfill cell as the previous studies. (6)

Table 2. The Effect of Centrifugation, MF, and UF on BOD_5, COD, and TOC of Leachate

Experiment	BOD_5 mg/ℓ	COD, mg/ℓ	TOC, mg/ℓ
Raw Leachate	16	448	352
Centrifugation	15	474	156
MF	16	498	376
UF-R[1]	29	902	532
UF-P[2]	3	64	8
Gel Permeation[3]	-	-	-

[1]"UF-R" means the retentant of UF process.

[2]"UF-P" means the permeate of UF process.

[3]No results were obtained in this study.

Table 3. Results of UF Membrane Evaluation On Molecular Weight Cut-off

Run	Sample, mg/ℓ	Retentant, mg/ℓ	Permeate, mg/ℓ
Blue Dextran	200	388	0
NaCl	6,000	7,380	5,029

reduced to 200 mℓ) with the UF unit (the recirculation rate was set at 225 mℓ/min and applied pressure at 40 psig with N_2 pressure). The UF process has a noticeable effect on BOD_5, COD and TOC values. These three parameters were concentrated about twice initial values in the retentate, and reduced to almost negligible amounts in the permeate (see Table 2). Therefore, it could be concluded that by applying a stabilized leachate through the UF system, most of the organics will be collected in the retentate fraction. The fraction of organics in the leachate with a molecular weight lower than 500 is practically negligible.

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